Report No. BMI-1583 (Del.)

UC-25 Metals, Ceramics,
and Materials
(TID-4500, 18th Ed.)

Contract No. W-7405-eng-92

PROGRESS RELATING TO CIVILIAN APPLICATIONS
DURING JUNE, 1962

by

Russell W. Dayton
Ronald F. Dickerson

July 1, 1962

BATTELLE MEMORIAL INSTITUTE
505 King Avenue
Columbus 1, Ohio

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

TABLE OF CONTENTS

	<u>Page</u>
REPORTS RELATING TO CIVILIAN APPLICATIONS ISSUED DURING JUNE, 1962	5
A. REACTOR MATERIALS AND COMPONENTS (AEC-DRD)	A-1
High-Pressure High-Temperature Solid-State Studies	A-1
Irradiation Surveillance Program on Type 347 Stainless Steel	A-1
Friction and Wear of Sliding Surfaces in Sodium Environments	A-2
Creep Studies of Irradiated Structural Materials	A-3
High-Burnup Irradiation of Boron-Compound Dispersions	A-6
Solubility of Helium in Heavy Water	A-7
Reactor Flow Studies	A-9
B. STUDIES OF FUELS (AEC-DRD)	B-1
Irradiation Studies of Niobium-Uranium Alloys	B-1
Development of Uranium-Containing Thorium Compounds	B-2
A Study of Plutonium-Containing Fuel Alloys	B-3
Measurements of the Volatility of PuO ₂	B-5
Fission-Product Release From Refractory Fuels	B-5
Effects of Fabricational Variables on the Radiation Stability of Uranium Carbide	B-6
Irradiation Studies on U ₃ Si ₂	B-8
Effects of High Burnup on UO ₂ -CeO ₂ and UO ₂ -ZrO ₂ Fuels	B-9
Irradiation Studies on Uranium-Molybdenum Alloys With Minor Additions	B-10
D. GENERAL FUEL-ELEMENT DEVELOPMENT (AEC-DRD)	D-1
Fabrication of Cermet Fuel Elements	D-1
Development of Low-Cost Tubular Fuel Elements	D-2
Gas-Pressure Bonding of Ceramic Fuel Elements	D-3
Factors Affecting Pressure Bonding	D-5
G. PREPARATION OF UO ₂ SINGLE CRYSTALS (AEC-DR)	G-1
Growth of UO ₂ Single Crystals	G-1
Characteristics of Heat-Treated UO ₂ Crystals	G-2
H. RADIOISOTOPE AND RADIATION APPLICATIONS (AEC-DID)	H-1
Use of Intrinsic Radioactive Tracers for Process Control	H-1
Graft-Polymerization Studies	H-3
A Study of the Effect of Internal Irradiation on Catalyst Activity	H-4
Establishment of Test Parameters for Sealed Radiation Sources	H-5
J. MATERIALS DEVELOPMENT AND EVALUATION (HAPO)	J-1
Mechanical Properties of Zirconium Alloys	J-1
Thermal Conductivity and Electrical Conductivity of UO ₂	J-2
L. COATED-PARTICLE FUEL MATERIALS (AEC-DRD)	L-1
MATERIALS DEVELOPMENT PROGRAM	L-1
Uncoated Fuel-Particle Studies	L-1
Alumina-Coating Studies	L-1
Carbon-Coating Studies	L-5
Beryllia-Coating Studies	L-6
Additional Oxide-Coating Studies	L-7
Chemical Fabrication Studies	L-8
Graphite-Matrix Studies	L-8
Pressure-Bonding Studies	L-10
FUEL-IRRADIATION PROGRAM	L-11
Sweep Capsule CFP-S-3	L-11
Sweep Capsule CFP-S-10	L-11
Postirradiation Studies	L-13
Intermediate-Temperature Design Studies	L-16
Fission-Gas Analytical Studies	L-16

TABLE OF CONTENTS
(Continued)

	<u>Page</u>
SUPPORTING RESEARCH PROGRAM	L-17
Radiation-Damage Studies	L-17
Fission-Gas-Diffusion Studies	L-17
Fuel-Compatibility Studies	L-19
Coolant-Compatibility Studies	L-20
Thermal-Conductivity Studies	L-21
Evaluation of Commercial Products	L-22
Fission-Fragment-Induced Strain Studies	L-23
M. CORROSION STUDIES OF THE FLUORIDE-VOLATILITY PROCESS (ORNL)	M-1
Q. PROGRESS ON USAEC/AECL COOPERATIVE PROGRAM (SRO)	Q-1
Long-Term Creep-Rupture Program on SAP Alloys	Q-1
R. FISSION-PRODUCT DEPOSITION STUDIES (ORNL)	R-1
S. RADIATION-EFFECTS STUDY OF CANDIDATE FUEL MATERIALS FOR MGCR (GA)	S-1
U. GAS-COOLED REACTOR PROGRAM (AGN)	U-1
ZrO ₂ -UO ₂ Fabrication Studies	U-1
ZrO ₂ -UO ₂ Design Studies	U-2
Postirradiation Examination of Hastelloy X Cladding From AGN 1B-8T-1 Subassembly	U-3
W. PROCESS DEVELOPMENT AND EVALUATION OF PROPERTIES OF F-48 NIOBIUM ALLOY (LRL)	W-1
X. GAS-PRESSURE BONDING OF BERYLLIUM-CLAD ELEMENTS (ORNL)	X-1

REPORTS RELATING TO CIVILIAN APPLICATIONS
ISSUED DURING JUNE, 1962

- BMI-1580 "The Chemical Reactions of PuO_2 With Reactor Materials",
by Stan J. Paprocki, Donald L. Keller, and William M.
Pardue.
- BMI-1581 "Progress Relating to Civilian Applications During May,
1962", by Russell W. Dayton and Ronald F. Dickerson.

REACTOR MATERIALS AND COMPONENTS (AEC-DRD)

R. F. Dickerson

Research having the general objective of providing structural-materials and component support to the over-all Atomic Energy Commission reactor-development program is reported in this section. A brief study concerned with the solubility of helium in heavy water is reported this month only.

High-Pressure High-Temperature Solid-State Studies

A. P. Young, D. A. Vaughan, and C. M. Schwartz

Objective

To determine if ultrahigh pressure can be utilized to produce new materials.

Program Approach

Samples of oxide mixtures are reacted at elevated temperatures under normal and ultrahigh pressures in sealed platinum capsules. Reacted samples are examined visually and by X-ray diffraction to determine phase relationships.

Progress Reported Previously

Effects of high pressure on reactions of uranium oxide with other metal oxides have been investigated. The mixed oxides have been treated as ternary systems. The effect of varying oxygen content on phases formed at high pressure have been examined.

Work This Period

A topical report describing and interpreting the effects of high pressure on reactions between uranium oxide and other metal oxides is being prepared. A paper describing the girdle high-pressure device has been prepared for presentation at the annual ASME meeting in 1962.

Plans for Future Work

Work in this area has been terminated.

Irradiation Surveillance Program on Type 347 Stainless Steel

W. E. Murr, F. R. Shober, and R. F. Dickerson

Objective

To determine the changes occurring in the mechanical properties of AISI Type 347 stainless steel samples and reactor pressure tubes when subjected to fast-neutron irradiation at process-water temperature (120 F) and up to 750 F.

Program Approach

A total of 144 test specimens are being or have been irradiated in core positions of the ETR. This group includes 12 Charpy V-notch specimens, 24 subsize Izod specimens, and 54 each of subsize tensile and cyclic-strain fatigue specimens. Evaluation of the irradiation-induced changes will be based on results of mechanical tests made on specimens irradiated to integrated neutron fluxes between about 5.0×10^{21} and greater than 3.0×10^{22} nvt. The information thus gained will be used to help predict the safe operating life for in-pile loops constructed of this material.

The irradiation-induced property changes in Type 347 stainless steel reactor components are being investigated by determining the tensile properties of samples machined from a pressure tube which was removed from the J-10 position of the ETR after an exposure of approximately 7.0×10^{21} nvt.

Progress Reported Previously

The tensile properties of Type 347 stainless steel specimens irradiated in ETR process water over an exposure range of 5.7×10^{21} to about 1.2×10^{22} nvt were measured. The tests were performed at room temperature and 600 F upon specimens in the as-irradiated and irradiated-and-annealed (1 hr at 1800 F) conditions. The results of these tests were given in the previous monthly progress report, BMI-1581. It was noted that the mechanical properties of specimens irradiated at the two different exposure levels were quite similar, the chief differences being slight decreases in total elongation and increases in yield strength for the higher exposure specimens. Total elongation in tests at 600 F showed a greater decrease than that obtained at room temperature, a matter of significance since nearly all test loops operate at elevated temperatures. There were, however, considerable differences observed between unirradiated tensile specimens. Annealing the test specimens for 1 hr at 1800 F was sufficient to remove nearly all effects of fast-neutron irradiation. The response of the annealed material to subsequent irradiation is not known.

Work This Period

Work this period has been concerned chiefly with metallographic examination and specimen sampling for subsequent radiochemical analysis. This work is in progress and has not reached a logical reporting stage at this time. Cyclic-strain fatigue testing of irradiated Type 347 stainless steel specimens has been continued. A specimen having a total neutron exposure of about 1.0×10^{22} nvt received a total of over 250,000 cycles at a total strain of about 0.5 per cent without fracture.

Plans for Future Work

Eight capsules containing subsize tensile and cyclic-strain fatigue specimens will continue to receive irradiation in the ETR. Cycle-strain fatigue testing will continue. Metallographic examination of several irradiated and annealed specimens will be performed, and the total fast-neutron exposure to which the specimens were subjected will be determined.

Friction and Wear of Sliding Surfaces in Sodium Environments

J. W. Kissel and W. A. Glaeser

Objective

To develop general principles applicable to selection of materials and design of mechanical parts suitable for applications involving molten sodium at temperatures to around 1300 F.

Program Approach

The friction and wear between various materials resistant to corrosion in molten sodium are being investigated by means of a slow-speed stick-slip apparatus. This employs a ball sliding across a flat specimen, and the environment is controllable from high vacuum (5×10^{-6} mm of mercury absolute) to several pounds (gage) of any selected inert gas. Detailed descriptions of this apparatus can be found in a topical report, BMI-1405.

Progress Reported Previously

Improved friction and wear performance has generally been observed for metal and metal-alloy materials following the addition of molten sodium between the rubbing surfaces. Studies of the composition of surface films found before and after sodium additions proved that reaction products were largely responsible for the improved behavior. On the other hand, friction did not decrease between cemented carbide surfaces in the presence of sodium. In this case, however, thermodynamic considerations do not favor the formation of reaction products. The experiments referred to are described in detail in a topical report, BMI-1405, and in ASLE Paper No. 61AM2B-1 and ASME Paper No. 61-LUBS-16.

A sodium purification loop and molybdenum capsule system have been devised for determining reaction rates of oxygen in sodium with pure molybdenum. The self-welding behavior of sodium-lubricated surfaces of Type 304 stainless steel against itself, as determined with a stick-slip friction apparatus, indicates that the static breakaway friction is about doubled after a 31-day exposure at 600 F.

Work This Period

Sodium losses by distillation during a 10-hr exposure experiment to determine reaction rates amounted to about 1 per cent per hr — an amount not considered serious. A duplicate self-welding experiment is in progress, and sodium is being periodically added to the system to insure continuous immersion at 600 F.

Plans for Future Work

Experiments will be performed to compare the reaction rates of oxygen in sodium with pure molybdenum and oxidized molybdenum (MoO_3) for exposure periods of from 1/4 to 1 hr. The self-welding experiment in progress will be continued until a 30-day exposure has accumulated.

Creep Studies of Irradiated Structural Materials

F. R. Shober, J. D. Bray, and R. F. Dickerson

Objective

To determine radiation-induced change in A-286 stainless steel, Hastelloy X, and René 41 by postirradiation hardness, creep-rupture, and tensile tests at temperatures up to 1200 F, and to determine by creep-rupture tests the recovery from irradiation effects of A-286 stainless steel and Hastelloy X after postirradiation heat treatment.

Program Approach

Type A-286 stainless steel, Hastelloy X, and René 41 are to be irradiated to fast-neutron flux exposures of approximately 1×10^{20} nvt at about 1000, 1200, and 1600 F to determine the 10- and 100-hr rupture strengths. The tensile properties are to be determined at room temperature and at 600, 1000, and 1600 F after irradiation. In addition to these properties, the hardnesses are to be measured after irradiation. The effect of postirradiation heat treatment on the tensile and creep properties at elevated temperatures is to be investigated. A comparison of the results obtained will be made with those of unirradiated materials.

Progress Reported Previously

Four remotely operated and shielded creep machines were placed in operation, and tests have been performed at various stresses and at temperatures up to 1600 F. Post-irradiation hardnesses of 79 specimens were reported in BMI-1569. Tensile-machine

grips were made, and furnace temperature-gradient adjustments and calibrations were completed. Tensile testing was begun.

Work This Period

Additional creep-rupture tests were initiated and completed at 1600 F. Test results are shown in Table A-1.

TABLE A-1. STRESS-RUPTURE PROPERTIES OF IRRADIATED HASTELLOY X
AT ELEVATED TEMPERATURES

Specimen	Condition	Approximate Irradiation Temperature, F	Test Temperature, F	Stress, psi	Failure Time, hr
34HS	As irradiated	1600	1600	8,000	60.7
50HS	As irradiated	1600	1600	8,000	54.0
35HS	As irradiated	1600	1600	10,000	26.0
46HS	As irradiated and heat treated ^(a)	1600	1600	8,000	27.9
52HS	As irradiated	1600	1600	6,000	257.5
51HS	As irradiated	1600	1600	8,000	118.6
54HS	As irradiated	1600	1600	8,000	79.5
47HS	As irradiated and heat treated ^(a)	1600	1600	5,000	In test
36HS	As irradiated	1600	1600	6,500	In test
56HS	As irradiated	1600	1600	10,000	14.0

(a) Postirradiation heat treatment involved holding at 2150 F for 1 hr in argon followed by rapid cooling in argon.

Tensile testing of 51 specimens was completed. Processing of the data is not complete. Preliminary test results are shown in Tables A-2 and A-3.

Plans for Future Work

Creep-rupture testing and processing of tensile data will continue.

TABLE A-2. TENTATIVE TENSILE PROPERTIES OF IRRADIATED A-286 STAINLESS STEEL, HASTELLOY X, AND RENÉ 41

Specimen	Test Temperature, F	0.2 Per Cent Offset Yield Strength, psi	Ultimate Strength, psi	Uniform Elongation ^(a) , per cent
<u>A-286 Stainless Steel, As Irradiated</u>				
67L	80	120,000	175,500	21.1
43L	80	111,000	168,500	20.6
69L	600	103,500	156,500	17.2
46L	600	114,000	157,500	15.2
71L	1000	119,000	139,500	3.4
49L	1000	115,000 ^(b)	141,500	7.0
52L	1200	107,000	107,000	0.7
5AS	1200	107,500	109,500	1.0
6AS	1200	102,000	104,000	1.1
13AS	1200	104,000	108,500	3.8
14AS	1200	100,000	113,000	4.4
<u>A-286 Stainless Steel, As Irradiated and Postirradiation Heat Treated^(c)</u>				
13L	80	120,500	169,000	18.5
74L	80	117,000	170,000	19.8
14L	1000	114,000	143,500	8.3
73L	1000	113,500	143,500	9.6
51L	1200	102,000	110,500	1.6
79L	1200	105,500	109,500	1.4
<u>René 41, As Irradiated</u>				
1R	80	111,500	134,000	7.7
12S	80	121,000	139,000	5.4
4R	1000	101,500	134,000	7.5
8RS	1200	105,000	140,500	8.8
10RS	1200	107,000	135,500	6.7
<u>Hastelloy X, As Irradiated</u>				
41H	80	54,400	107,700	>36.0
21H	80	59,900	112,500	>36.0
37HS	80	43,800	111,400	31.2
44H	1000	32,900	82,500	>36.0
46H	1000	42,000	90,600	34.4
1HS	1200	31,900	73,700	22.4
2HS	1200	32,500	73,800	20.4
41HS	1200	33,100	85,000	22.5
32HS	1600	26,000	37,500	3.6
39HS	1600	25,000	35,500	3.8
<u>Hastelloy X, As Irradiated and Postirradiation Heat Treated^(c)</u>				
29H	80	44,100	108,300	>36.0
43H	80	43,900	108,100	>36.0
40H	1000	30,900	88,300	>36.0
72H	1200	29,500	76,000	33.0
3SH	1200	29,000	77,000	34.0
42HS	1600	26,500	37,500	3.6
45HS	1600	28,000	39,000	3.3

(a) Elongation at maximum load.

(b) Equipment malfunction during test; value estimated.

(c) Postirradiation heat treatments: A-286 stainless steel: 2 hr at 1650 F, oil quenched; 16 hr at 1325 F, inert gas cooled; 16 hr at 1200 F, inert-gas cooled. Hastelloy X: 1 hr at 2150 F, inert-gas cooled.

TABLE A-3. TENSILE STRENGTH OF NOTCHED IRRADIATED TENSILE SAMPLES
AT 1000, 1200, and 1600 F

Specimen	Material	Test Temperature, F	Breaking Load, lb	Ultimate Tensile Strength ^(a) , psi
9AN	A-286	1000	2210	197,000
10AN	A-286	1200	1025	82,000
15AN	A-286	1200	880	70,500
16AN	A-286	1200	959	76,600
17AN	A-286	1200	1706	136,500
18AN	A-286	1200	1700	136,000
9HN	Hastelloy X	1200	1065	85,300
10HN	Hastelloy X	1200	1240	99,000
19HN	Hastelloy X	1600	494	39,400
18HN	Hastelloy X	1600	625	50,000
7RN	René 41	1200	1910	153,000
8RN	René 41	1200	1975	158,000

(a) The root diameter of all notched specimens is nominally 0.125 in.

High-Burnup Irradiation of Boron-Compound Dispersions

R. J. Burian and J. E. Gates

Objective

To study the effect of burnup of 80 per cent of the boron-10 in dispersions of B_4C in Al_2O_3 , B_4C in Zircaloy-2, and ZrB_2 in Zircaloy-2 at temperatures in the region of 400 to 650 F.

Program Approach

The Martin-Marietta Company is developing neutron poisons for use in advance PM-1 type reactors. Several materials being considered for use as poisons in these reactors will be irradiated in this program to determine the effects of high burnup of boron-10. Dispersions of B_4C in Al_2O_3 , B_4C in Zircaloy-2, and ZrB_2 in Zircaloy-2 are involved. Specimens of the poison materials will be irradiated under conditions of size, composition, boron burnup, temperature, and cladding as near those of the prototype as possible.

The irradiations will be conducted in three capsules, each containing 20 pellets of the selected poisons. The pellets will be of prototype size, nominally 0.460 in. in diameter by 0.50 in. long, containing either 0.017 or 0.025 g of boron-10 per pellet. The pellets in each capsule will be divided into four groups of five pellets with each group to be separately contained inside sealed stainless steel tubes. The capsules will be equipped with electrical heaters and thermocouples. Irradiation of the capsules will be conducted in the ETR to achieve burnups of about 80 per cent of the boron-10 at temperatures of 400 to 500 F at the surface of the pellets.

The effects of variations in particle size, pellet density, and fabrication methods on the helium-retention properties, dimensional stability, mechanical stability, and corrosion properties of the poison materials will be evaluated after irradiation. Microstructural studies will also be performed, and the amount of boron-10 burned during irradiation will be determined by chemical analyses.

Progress Reported Previously

Three capsules identified as MND-2-1, MND-2-2, and MND-2-3 were designed and fabricated. The capsule design was described in BMI-1577.

Twelve specimens were fabricated from ten different lots of poison pellets. These ten lots of pellets are identified in BMI-1577 as to the specimen identification and the capsule in which the specimens will be irradiated.

Results of quantitative metallographic examinations indicated that a relatively inhomogeneous dispersion of the poison particles within the pellets existed such that the density of particles varied by a factor of 2 or more at different locations in the pellets. The results of analyses of the material were presented last month in BMI-1581 with the results of analyses performed by the Martin-Marietta Corporation and by the Carborundum Company. These data showed good correlation.

Assembly of the three irradiation capsules was completed. The units were shipped to the ETR for irradiation.

Work This Period

The capsules were accepted for irradiation after a routine preinsertion check conducted by MTR personnel. The irradiation is tentatively scheduled to begin in July, 1962.

Plans for Future Work

The irradiation will be conducted for three ETR cycles to a burnup of approximately 80 per cent of the boron-10 atoms. The specimens will then be examined and evaluated.

Solubility of Helium in Heavy Water

E. F. Stephan and W. E. Berry

Objective

To determine the solubility of helium in heavy water (D_2O) at 50, 100, and 150 C at total pressures in the range of 700 psi and at 200, 240, and 280 C in the ranges of 1000 and 1300 psi.

Program Approach

The technique and equipment used for obtaining the solubility data, described in detail in BMI-1067, are briefly summarized as follows: From a 1-liter pressure vessel at temperature and pressure, a sample of the liquid is expanded into a thermostatically cooled, mercury-filled, gas burette, and the pressure is adjusted to atmospheric. The volume of the gas and the liquid are measured and adjusted to standard conditions. Equilibrium between gas and liquid is assumed, when successive samples taken 2 hr apart show no change in gas content.

Progress Reported Previously

The program was initiated this month.

Work This Period

The solubility of helium in heavy water was determined at 50, 100, 150, 200, 240, and 280 C for helium partial pressures ranging from 50 to 1150 psi. The averaged data for several successive samples taken at equilibrium are listed in Table A-4. The solubility at temperature appears to be a linear function of the partial pressure of the helium. The plotted curve of solubility versus partial pressure is a straight line passing through

the origin. The calculated Henry's law constants for the various temperatures are also included in Table A-4.

TABLE A-4. SOLUBILITY OF HELIUM IN HEAVY WATER (D₂O)

Temperature, C	Partial Pressure of Helium, psia	Solubility, ml He, STP, /g D ₂ O	Henry's Law Constant ^(a) , K x 10 ⁵
50	668	0.437	17.12
100	723	0.559	14.49
150	640	0.711	10.08
200	788	1.26	6.99
	1093	1.72	
240	506	1.31	4.44
	797	2.02	
280	77	0.31	2.84
	377	1.56	

$$(a) K = \frac{1120 P(\text{pp He})}{\text{ml He, STP, /g D}_2\text{O}} .$$

The solubility of helium in light water was determined at several temperatures and pressures for comparison purposes. These data are listed in Table A-5. Comparison of the light- and heavy-water data indicates that the solubility of helium in D₂O

TABLE A-5. SOLUBILITY OF HELIUM IN LIGHT WATER (H₂O)

Temperature, C	Partial Pressure of Helium, psia	Solubility, ml He, STP, /g H ₂ O	Henry's Law Constant ^(a) , K x 10 ⁵
200	395	0.75	
	415	0.79	
	785	1.47	6.65
	805	1.48	
	835	1.53	
240	475	1.34	
	495	1.39	
	515	1.43	4.41
	615	1.77	
	645	1.79	
	715	2.05	
280	177	0.85	
	217	1.03	
	257	1.19	2.52
	337	1.72	
	357	1.75	

$$(a) K = \frac{1244 P(\text{pp He})}{\text{ml He, STP, /g H}_2\text{O}} .$$

is the same as in H_2O on a volume basis. A good approximation of the solubility of the gas in D_2O can be obtained by multiplying the solubility in H_2O by the ratio of the densities (approximately 0.9) of the two types of water.

Plans for Future Work

The programmed experimental work has been completed. A topical report will be prepared.

Reactor Flow Studies

L. J. Flanigan and H. R. Hazard

Objective

To study coolant flow and mixing in the PWR Core 2.

Program Approach

A 0.4-scale model, using air as the working fluid, is being used to obtain data for the design of the prototype.

Progress Reported Previously

Model studies of flow and mixing with four loops, three loops, two adjacent loops, and two opposed loops operating were finished. A topical report covering all of the flow studies was drafted.

Work This Period

The draft of the topical report is being reviewed.

Plans for Future Work

The program has been concluded.

STUDIES OF FUELS (AEC-DRD)

R. F. Dickerson

Research concerned with providing fuel-material support for the over-all reactor-development program of the Atomic Energy Commission is reported in this section. A study concerned with the postirradiation examination of uranium-molybdenum alloys with minor ternary additions has been initiated and is reported for the first time.

Irradiation Studies of Niobium-Uranium Alloys

J. A. DeMastry, A. A. Bauer, and R. F. Dickerson

Objective

To investigate the irradiation stability of high-strength niobium-uranium alloys.

Program Approach

A program has been in progress to investigate and determine those physical, mechanical, and corrosion properties of niobium-uranium alloys which are of interest in evaluating their usefulness as reactor fuels. These studies have been completed, and the irradiation behavior of these alloys is now being investigated. For this purpose, two capsules containing 15 specimens each were irradiated to a maximum burnup of 1 total a/o at intended temperatures of 1600 and 1800 F. The specimens include binary niobium alloys containing 10, 20, and 30 w/o uranium and ternary alloys containing 20 w/o uranium with 10 and 20 w/o zirconium.

Progress Reported Previously

Property determinations related to this program have been completed and were discussed in two topical reports, BMI-1400 and BMI-1536. The planning of and construction of capsules for irradiation tests were reported in BMI-1464.

The first capsule, WR-20, was irradiated in a core position in the WTR for six cycles. This capsule operated for one cycle at 1600 F and between 1200 and 1300 F for the remaining cycles.

Capsule WR-21 operated for three cycles at 1800 F. At the end of the third cycle the WTR was shut down, and Capsule WR-21 was removed. Both capsules were returned to the BMI Hot-Cell Facility.

Both capsules were opened, and visual examinations and stereomacrographs were made of each specimen. Alloys containing 10 and 20 w/o uranium appeared to be physically sound; some of those containing 30 w/o uranium showed some surface roughness; while those containing zirconium additions exhibited surface cracking. A summary of the dimensional changes observed was given last month in BMI-1581.

Work This Period

An analysis of temperature data from capsule thermocouples indicates that certain specimens in Capsule WR-21 attained temperatures of 1900 F while most specimens in Capsule WR-20 experienced temperatures between 1100 and 1600 F.

Plans for Future Work

All experimental work including bend tests and metallographic examinations of irradiated samples has been completed. The data are being analyzed. Preparation of a topical report has begun.

No further experimental work is planned.

Development of Uranium-Containing Thorium Compounds

J. A. DeMastry, A. A. Bauer, and R. F. Dickerson

Objective

To investigate thorium-uranium compounds with the aim of developing high-temperature fuels for thermal breeder reactors and of determining those properties which will indicate the suitability of such compounds for various reactor applications.

Program Approach

Thorium-uranium aluminides, beryllides, borides, carbides, nitrides, silicides, and sulfides were selected as promising materials for study using high melting point, low thermal-neutron cross section, high thorium (and uranium) density, and crystal structure as the criteria for selection. These compounds are being evaluated as to preparation, corrosion resistance in various media, and mechanical and physical properties. The four most promising compounds were selected for more detailed study, primarily on the basis of ease of preparation by casting as determined during the first year of study. These compounds and compositions were $(Th_9U)B_4$, $(Th_9U)C$, $(Th_9U)C_2$, and $(Th_9U)Be_{13}$.

Progress Reported Previously

During the first phase of this program, data were obtained on thorium and thorium-uranium compounds of aluminum, beryllium, boron, carbon, and silicon. The results of these studies are reported in BMI-1568.

The results of thermal-expansion measurements from 75 to 1800 F and of compression tests at 75 and 1200 F on specimens of $(Th_9U)B_4$, $(Th_9U)C$, and $(Th_9U)C_2$ were reported in BMI-1569. No evidence of ductility was observed in the compressive-strength tests.

Oxidation-test results for the four compounds of interest also were reported in BMI-1569 and in BMI-1574. The results indicated that the $(Th_9U)B_4$ compounds exhibited exceptional oxidation resistance at temperatures up to 2500 F. Microstructural examination of the $(Th_9U)C$ compound specimens which were oxidized at 1100 and 1300 F for 15 and 30 min showed no evidence of intergranular penetration by oxygen.

Heat treatments of the $(Th_9U)C$, $(Th_9U)C_2$, and $(Th_9U)B_4$ compounds in contact with niobium, tantalum, stainless steel, and zirconium for 10 hr at 1000 and 1500 F and in contact with niobium, tantalum, and tungsten for 10 and 100 hr at 2000 and 2500 F were completed.

Work This Period

All packs for compatibility studies were opened and examined except one which was annealed for 100 hr at 2500 F. Bonding between the compounds and potential cladding material was not achieved, although bonding between adjacent disks of cladding material was observed. No evidence of attack of the cladding by the compounds was noted. Bonding is apparently being prevented by an oxide coating or some type of surface film on the compounds. Additional attempts to achieve bonding by compressive loading at elevated temperatures in a tensile machine will be attempted.

Additional compression tests were attempted at 2000 F for the $(Th_9U)C$ compound. At this temperature a limited amount of ductility can be expected in the compound, reflected in higher fracture strengths than are observed at lower temperatures. The yield strength of the compression jig was exceeded with a stress of 56,000 psi on the specimens, and the tests had to be discontinued. The specimens did not fracture. Future work at higher temperatures will require a jig made of higher strength materials as well as cooling of the jig to minimize deformation.

Casting of the $(\text{Th}_9\text{U})\text{Be}_{13}$ compound has been delayed, and the next phase of the study has been initiated. The preparation of the compounds Th_3Si_2 , ThSi , ThN , Th_2Bi , and ThS is being investigated by arc-casting methods.

Thermal-conductivity measurements of the $(\text{Th}_9\text{U})\text{C}$, $(\text{Th}_9\text{U})\text{C}_2$, and $(\text{Th}_9\text{U})\text{B}_4$ compounds were delayed by equipment failures. These tests have been resumed and are currently in progress.

Plans for Future Work

Thermal-conductivity measurements will be completed. Compatibility studies of the compounds of interest will be continued.

The $(\text{Th}_9\text{U})\text{B}_{13}$ compound will be prepared and the properties determined as soon as possible. When castings of the Th_3Si_2 , ThSi , ThN , Th_2Bi , and ThS compounds are available, oxidation tests and compressive-strength measurements will be made to evaluate the compounds.

A Study of Plutonium-Containing Fuel Alloys

V. W. Storhok, A. A. Bauer, and R. F. Dickerson

Objective

To investigate those plutonium alloys which appear to have potential as fuels for relatively high-temperature propulsion applications or for possible use as "spike" fuel elements in power reactors.

Program Approach

Plutonium-containing alloy systems have been selected for study primarily on the basis of the behavior of similar uranium-alloy systems. The compositions selected are based on the iron, niobium, thorium, and uranium-niobium systems. These systems are being studied at the plutonium-lean end. Initial evaluation is based on metallographic studies and hardness measurements. Those alloys exhibiting desirable phase distribution and evidence of strength on the basis of hardness measurements will be subjected to more detailed property studies.

Progress Reported Previously

Studies of cast dispersion fuel alloys with iron or niobium matrices are in progress.

Iron-27.6 w/o plutonium, iron-27.6 w/o plutonium-11.9 w/o chromium, and iron-27.6 w/o plutonium-2.1 w/o molybdenum alloys were found to have cast structures consisting of iron or iron alloy surrounded by a eutectic mixture of PuFe_2 and iron (BMI-1561). Heat treating the iron-plutonium and iron-plutonium-molybdenum alloys for up to 20 hr at a nominal temperature of 900 C resulted in a coalescence of the PuFe_2 phase into discrete particles occupying approximately 30 volume per cent of the alloy (BMI-1565). In the as-cast condition, the iron-base alloys were capable of sustaining reductions in thickness of 20 to 25 per cent by rolling at 900 to 950 C (BMI-1577). Heat treating the alloys for 4 hr at 900 C did not appreciably alter the fabrication characteristics. The alloys apparently cannot be cold rolled. On the basis of one differential thermal-analysis run, the solidus temperature of the iron-27.6 w/o plutonium-11.9 w/o chromium alloy is 1148 C (BMI-1581).

The cast structure of a niobium-25.6 w/o plutonium-6.1 w/o silicon alloy consists of niobium and approximately 30 volume per cent PuSi_2 as a semicontinuous phase (BMI-1549). A heat treatment of 20 hr at 1000 C resulted in the coalescence of the PuSi_2 phase

into discrete particles (BMI-1577). The heat-treated alloy exhibited enough ductility during rolling at 550 C to suggest that significant reductions could be made at higher temperatures (BMI-1581).

Uranium-niobium alloys are also being studied as possible breeder-type fuels. Structures of uranium-10 w/o niobium-5, -10, -15, and -20 w/o plutonium alloys consist essentially of gamma-uranium solid solution after a heat treatment of 10 hr at 900 C followed by an oil quench. An apparent second phase in the 20 w/o plutonium alloy may be caused by coring (BMI-1581). Alloys furnace cooled from 900 C consist of gamma-uranium solid solution and a transformation product believed to be alpha uranium and/or zeta uranium-plutonium. Hardnesses of the as-cast, oil-quenched, and furnace-cooled alloys increased with increasing plutonium content (BMI-1574 and BMI-1581).

Work This Period

Several additional dispersion-type alloys have been prepared for study by arc-melting techniques. These include alloys intended to consist of 20 and 25 volume per cent PuFe_2 in iron and iron-20 w/o chromium matrices and 20 and 25 volume per cent PuSi_2 in niobium matrices. Stainless steel and niobium packs are being prepared for an investigation of the pack-rolling fabrication characteristics of these alloys and of alloys containing 30 volume per cent of dispersed compound phase.

Metallographic examination of hot-rolled iron-27.6 w/o plutonium, iron-27.6 w/o plutonium-11.9 w/o chromium, and iron-27.6 w/o plutonium-2.1 w/o molybdenum alloys (alloys containing 30 volume per cent PuFe_2) revealed a slight coalescence of the PuFe_2 phase after a heat treatment of 20 hr at 900 C. About the same degree of coalescence of PuFe_2 was noted after the worked alloys were heat treated for 4 hr at 1000 C. There is strong evidence that the heat-treatment temperature of as-cast iron-base alloys in another furnace was actually closer to 1000 C rather than 900 C as previously reported. An incorrectly positioned thermocouple is believed responsible for the error.

Averaging the results of two differential thermal-analysis runs gives a solidus temperature of 1162 C for the iron-27.6 w/o plutonium alloy. On the basis of one run, the solidus temperature of the iron-27.6 w/o plutonium-2.1 w/o molybdenum alloy is 1159 C. Additional thermal analysis of these alloys is necessary to confirm these data.

Microstructures of uranium-10 w/o niobium-5, -10, -15, and -20 w/o plutonium alloys heat treated 10 hr at 900 C, oil quenched, and aged 2 hr at 500 C consist of gamma uranium and a dispersed phase believed to be alpha uranium and/or zeta uranium-plutonium. Hardnesses of these alloys in this condition of heat treatment were 369, 449, 549, and 520 DPH for the 5, 10, 15, and 20 w/o plutonium alloys, respectively.

Plans for Future Work

The effects of reduced plutonium-compound content and of pack rolling on the fabricability of iron-base and niobium-plutonium-silicon alloys will be investigated.

Additional attempts will be made by differential thermal analysis to confirm the previously reported solidus temperatures of the iron-base alloys and to establish the solidus temperatures of thorium-20 and -30 w/o plutonium alloys.

Studies of the effects of heat treatment on the structures of iron-plutonium, niobium-plutonium-silicon, and uranium-niobium-plutonium alloys will continue.

Measurements of the Volatility of PuO₂

S. J. Paprocki, D. L. Keller, C. A. Alexander, and W. M. Pardue

Objective

To determine the volatility of PuO₂ in the 1400 to 1750 C temperature range.

Program Approach

The volatility of PuO₂ will be determined in various atmospheres. Transpiration vapor-pressure techniques will be utilized with analysis of the condensate accomplished by absolute alpha-radiation counting methods. On the basis of an abbreviated prior study, it is predicted that PuO₂ will be the congruently vaporizing plutonium-oxide compound in nonreducing atmospheres. The vapor pressures are expected to be several orders of magnitude lower than those of uranium oxides under oxidizing conditions. X-ray diffraction studies will be performed subsequent to some of the transpiration runs in an attempt to determine the elevated-temperature structure of the oxides. Such interpretation of room-temperature studies may be justified on the basis of the relatively sluggish phase transformations generally associated with oxide systems.

Progress Reported Previously

Transport rates have been measured and determined to be temperature independent. The mobility is attributed to alpha self-propulsion and is sufficiently high to require compensation during vapor-pressure calculations. Gas flow is conducive to the particulate mobility.

The variation in vapor pressure as a function of temperature has been determined in air at 1450, 1500, 1555, 1600, 1650, 1700, and 1775 C with an essentially linear relationship between temperature and vapor pressure resulting. The range of vapor pressures was between 1.09×10^{-9} atm at 1450 C and 2.95×10^{-8} at 1775 C.

Numerous additional runs have been performed in oxygen and argon as well as air atmospheres in the temperature range of interest. Reproducibility of results has been demonstrated.

One volatility determination was performed in a hydrogen atmosphere. In addition, X-ray diffraction studies of the residue after experiments have been presented.

Work This Period

No experiments were performed during the past month due to an equipment failure. Writing of a final report covering the entire program and including a proposed mechanism of vaporization has been initiated.

Plans for Future Work

Program has been concluded.

Fission-Product Release From Refractory Fuels

J. B. Melehan and F. A. Rough

Objective

To establish an understanding of the important causes of fission-product release during irradiation of refractory fuels, starting with uranium dioxide, and to obtain engineering information which will aid in the design of fuel elements utilizing these fuels.

Program Approach

The present program consists of experiments to study the fission-gas-release behavior of UO_2 during irradiation. UO_2 samples of densities between 90 and 100 per cent of theoretical are being irradiated in the BRR at temperatures to 3000 F. Fission gases released during irradiation are periodically collected and analyzed for isotopic release rates by gamma spectrometry.

It has been possible to determine release rates for krypton and xenon isotopes over a wide range of half-lives (from krypton-92 with a 3-sec half-life to xenon-133 with a 5.26-day life) and hence to test how fission-gas-release properties depend on half-life. It has also been possible to obtain information on the release characteristics of iodine-133 and iodine-135 by measuring the residual xenon-133 and xenon-135 release rates following reactor shutdown.

Progress Reported Previously

The irradiation of high-density (98 per cent of theoretical) sintered UO_2 was begun at 1800 F. The experiment was concluded because the release rate showed an unexpected increase after having been at a steady-state value. Examination of the UO_2 sample for structural changes was planned but not completed.

Work This Period

The irradiation of high-density sintered UO_2 was resumed this month. The irradiation sample has the same density as had the sample in the discontinued 1800 F irradiation. Irradiations at 2000, 2300, 2500, and 2700 F, each of 48 to 72 hr in duration, have been completed. Fission-gas samples have been analyzed for abundance of krypton-85m, xenon-133, and xenon-135. Fission-gas release rates have not yet been calculated. Several daughter-trap samples have been obtained for analysis in an attempt to determine the release rates of fission gases with half-lives of several minutes or less. Using release-rate values for isotopes possessing a wide range of half-lives, it will be possible to establish the relation between half-life and release rate and consequently establish the principal mechanism of gas release at several temperatures.

Plans for Future Work

The program of high-temperature UO_2 irradiations will continue with additional exposures at 2500, 2700, and 3000 F. The 2500 F test will be a repetition of the already completed 2500 F exposure to determine whether the 2700 F irradiation caused any structural changes in the UO_2 sample. An attempt will also be made to determine release rates for iodine-133 and iodine-135 precursors.

Effects of Fabricational Variables on the Radiation Stability of Uranium Carbide

D. G. Freas, E. O. Fromm, J. F. Lagedrost,
J. H. Stang, J. E. Gates, and R. F. Dickerson

Objective

To investigate the effects of compositional and fabricational variables on the radiation stability of uranium monocarbide at irradiation temperatures of about 1380 F at the fuel surface, fission-heat-generation rates of about 1.5×10^6 Btu/(hr)(ft²), and fuel burnups of about 15,000 MWD/TU.

Program Approach

Five uranium carbide specimens will be irradiated in each of three instrumented capsules in the MTR as part of the United Nuclear Corporation fuel-cycle program. These specimens will be fabricated by United Nuclear by both arc-casting and powder-metallurgy techniques. The carbon contents will range from approximately 4.4 to 5.2 w/o, and the uranium will be of 12 per cent enrichment. The fuel specimens will be 3/8 in. in diameter by 3/4 in. long and will be contained in niobium-1 w/o zirconium cans with 30-mil-thick walls. The fuel specimens in the first two capsules will include a liquid-sodium bond between the cladding and fuel. For the third capsule, specimens will be fabricated with a mechanical bond between the cladding and fuel.

The anticipated irradiation conditions for these specimens will include surface temperatures of 1380 F and center-line temperatures of 2280 F. The target irradiation exposure is 15,000 MWD/T burnup of the uranium. To evaluate the effects of the compositional and fabrication variables, the postirradiation examinations will include measurements of physical properties and fission-gas release and studies of microstructure.

Progress Reported Previously

Measurements of physical properties and microstructural studies of fuel specimens produced by the United Nuclear Corporation for this irradiation program have been completed. Fabrication of the irradiation capsules as described in BMI-1534, assembly of both the sodium- and mechanical-bonded specimens as described in BMI-1565, and assembly of the three capsules, UNC-1-1, UNC-1-2, and UNC-1-3, have been completed. The three capsules are now being irradiated in the MTR.

Capsule UNC-1-1. Capsule UNC-1-1 was inserted in MTR Position A-39-SW for Cycle 171. During Cycles 171 and 172, the specimen-surface temperatures ranged from 1425 to 1600 F with center-line temperatures ranging from 2325 to 2500 F. At the end of Cycle 174 on June 4, 1962, this capsule will have been irradiated for about 62 days.

Capsule UNC-1-2. The irradiation of Capsule UNC-1-2 was initiated during Cycle 170 in MTR Position A-38-NE. However, the capsule was moved to A-38-NE and then to A-39-SE in order to achieve the desired specimen-surface temperature. During Cycles 171 and 172 the surface temperature of the specimens ranged from 1325 to 1500 F, with center-line temperatures ranging from 2225 to 2400 F. At the end of Cycle 174, this capsule will have been irradiated for about 80 days.

Capsule UNC-1-3. Capsule UNC-1-3 was inserted into MTR Position A-7-SW for Cycle 172. During that cycle, the specimen-surface temperatures ranged from 1350 to 1525 F, and the center-line temperatures ranged from 2250 to 2425 F. At the end of Cycle 174, this capsule will have been irradiated for about 48 days.

It has been estimated that design burnups of 15,000 MWD/T will require approximately 135 days for each experiment. This is a somewhat longer in-pile time than was originally estimated. The increased time required reflects the fact that fluxes are probably somewhat lower than those estimated in the original capsule-design studies.

Work This Period

Irradiation of the three capsules was continued. Estimated fuel-surface temperatures, based on thermocouple data, were in the range 1300 to 1600 F. Auxiliary electric heat was used in UNC-1-2 and UNC-1-3 to maintain the specimen temperatures.

New estimates were made of the in-pile time required to reach the 15,000-MWD/T burnup goal for each capsule. These estimates indicate that approximately ten MTR cycles will be required for each capsule. The discharge dates on this basis are:

<u>Capsule</u>	<u>Tentative Discharge Date</u>
UNC-1-1	October 8, 1962
UNC-1-2	October 8, 1962
UNC-1-3	October 29, 1962

The irradiation of Capsule UNC-1-1 may be extended until the specimens achieve a burnup of 30, MWD/T. Present estimates indicate that this burnup will be reached by July, 1963, assuming that the present irradiation conditions are maintained.

Plans for Future Work

The irradiation of the three capsules containing UC specimens will be completed and postirradiation examinations performed. An evaluation of the irradiated UC specimens will then be performed to determine the effects of fabrication variables, if any, on the radiation stability of the fuel.

Irradiation Studies on U_3Si_2

M. Kangilaski, J. E. Gates, and R. F. Dickerson

Objective

To determine the condition of U_3Si_2 specimens after irradiation at 1200 F to a burnup of about 10,000 MWD/TU.

Program Approach

Six 0.35-in. -diameter U_3Si_2 fuel rods bonded with sodium to a stainless steel cladding were irradiated under the supervision of AI personnel in a single capsule. Proposed burnup of these specimens was 10,000 MWD/TU and the specimen-surface temperature was expected to be about 1200 F. After irradiation, these fuel rods were shipped to Battelle for postirradiation examination. The examination will consist of length, diameter, and density measurements, gamma scanning, burnup analyses and metallographic examination of both the stainless steel cladding and the U_3Si_2 specimens. The fission gas released from the fuel and possible silicon migration from the fuel will also be determined.

Progress Reported Previously

The capsule was gamma scanned and the gases contained inside the capsule were sampled for analysis. Measurements of the specimens indicated diametral changes ranging from 2.4 to 6.3 per cent, length changes ranging from 1.8 to 4.2 per cent, and decreases in density ranging from 6 to 7 per cent. Metallographic examination of the specimens showed the presence of a second phase which was thought to be U_3Si . This phase appeared to be more prominent at the edge of the specimens. The microstructure of the Type 304 stainless steel cladding did not appear changed by the irradiation.

Work This Period

Results of chemical analyses of the specimens revealed that two of the specimens, 7 and 14, had lost a large amount of silicon. It was observed that the specimens which experienced the largest loss of silicon were those in which the second phase was observed during the metallographic examination. These results reinforce the theory that the second phase was U_3Si .

The sodium from the capsules was analyzed and found to contain 0.5 w/o silicon. Silicon analyses of the cladding were inconclusive because silicon is present as an original constituent in Type 304 stainless steel.

Plans for Future Work

Attempts will be made by X-ray diffraction techniques to positively identify the second phase which was found. Also, the fission-gas release will be correlated to the analytically determined burnup.

Effects of High Burnup on $\text{UO}_2\text{-CeO}_2$ and $\text{UO}_2\text{-ZrO}_2$ Fuels

J. E. Gates, R. A. Wullaert, E. O. Fromm, and J. H. Stang

Objective

To investigate the ability of homogeneous ceramic fuels to withstand burnups of up to 70 per cent of the uranium at surface and center-line temperatures of about 600 and 4000 F, respectively.

Program Approach

The irradiation stability of stainless steel-clad solid-solution ceramic fuels fabricated as part of a fuel-development program at Combustion Engineering will be investigated after fuel fission burnups of 40, 55, and 70 a/o. The fuels will consist of highly enriched UO_2 in CeO_2 or in ZrO_2 stabilized with CaO . Up to 24 cylindrical specimens will be irradiated in six thermocoupled capsules. An attempt will be made to sustain the target temperature levels throughout the irradiation by relocating the capsules into progressively higher neutron fluxes.

The postirradiation examination will involve detailed visual examination, dimensional and density measurements, metallographic and X-ray diffraction examinations, a fission-gas release study of selected specimens, burnup determinations by dosimetry and isotopic analyses, and corrosion testing of selected specimens in 650 F water.

Progress Reported Previously

An irradiation-parameter study was conducted which consisted of determining the interdependencies among fuel composition, target fuel conditions, burnup rates, neutron fluxes, and specimen diameters. Various details relative to the study were presented in BMI-1581. Nuclear mock-up experiments were planned to assess the validity of pertinent flux-perturbation relationships.

Work This Period

The irradiation-parameter study was continued. Values of specimen-effective (fuel core plus Type 347 stainless steel cladding) thermal-conductivity data provided to Battelle by CE are presented in Table B-1. Fuel-core diameters developed from these thermal conductivities are presented in the table. These sizes will be adopted for the irradiations unless data from the nuclear mock-up experiments indicate some need for adjustment. The latter experiments, which are currently being conducted at the Battelle Research Reactor, are designed to provide information relative to variations in neutron perturbation as burnup progresses. Interpretation of the mock-up data in terms of absolute perturbation values is not a primary objective because of the difficulty of accounting for the effect of the BRR core geometry as opposed to the MTR or ETR core geometry.

TABLE B-1. CONSTANTS FOR IRRADIATION STUDY OF $\text{UO}_2\text{-CeO}_2$ AND $\text{UO}_2\text{-ZrO}_2$ FUELS

Fuel	Composition	Effective Thermal Conductivity, Btu/(ft)(hr)(F)	Core Diameter Based on This ^(a) Effective Thermal Conductivity, in.
Z-10	10 w/o UO_2 ^(b) -9 w/o CaO-81 w/o ZrO_2	1.01	0.35
C-10	10 w/o UO_2 -90 w/o CeO_2	1.56	0.32
Z-22	22 w/o UO_2 -7.8 w/o CaO-70.2 w/o ZrO_2	0.80	0.25
C-25	25 w/o UO_2 -75 w/o CeO_2	0.82	0.19

(a) The core diameters are also based on specimen cladding-to-fuel core temperature gradients as follows:

Fuel	Temperature, F
Z-10	3400
C-10	2800
Z-22	3400
C-25	2800

(b) All UO_2 is highly enriched.

In addition to the analytical work directed toward establishing leading irradiation parameters, capsule design and fabrication of some components were initiated. The capsule design consists of a double-wall finned-annulus configuration with sodium as the thermal bond between the specimen cladding and the inner capsule shell. Two stainless steel-sheathed thermocouples will be attached to the cladding at the midpoint of each fueled sample for temperature monitoring.

Plans for Future Work

Any adjustments in specimen diameter deemed advisable from the nuclear mock-up experiments will be made. Fabrication of capsule components will continue.

According to the present plan, the first two capsules, designated BMI-43-1 and BMI-43-4, will be assembled during August and delivered to the reactor during September. These capsules will contain samples scheduled for 40 per cent fission burnup; the former will contain Z-10 and C-10 samples (see Table B-1) while the latter will contain Z-22 and C-25 samples. The other four capsule systems will be assembled during September and early October.

Irradiation Studies on Uranium-Molybdenum Alloys With Minor Additions

M. Kangilaski, J. E. Gates, and R. F. Dickerson

Objective

To determine the stability of uranium-molybdenum alloys with minor alloying additions during irradiations at 400 and 500 C to burnups of about 10,000 MWD/TU.

Program Approach

Four capsules containing a total of 64 specimens were irradiated under the supervision of AI personnel. Proposed burnup of these specimens was 10,000 MWD/TU at specimen-surface temperatures of 400 and 500 C. The specimens were plates (0.4 by

0.3 by 0.075 in.) and contained 3.5, 5, 7.5, and 10 w/o molybdenum. Some of the specimens also contained minor amounts of aluminum, niobium, and silicon. After irradiation these specimens were shipped to Battelle for examination. The examination will consist of length, width, thickness, microhardness, and density measurements, gamma scanning, burnup analyses, and metallographic examinations.

Progress Reported Previously.

Work on the program has just been initiated.

Work This Period

All four of the capsules were opened, the NaK was reacted, and the specimens were removed. Dimensions of the specimens were measured. These measurements were complicated by considerable swelling and bowing of some of the specimens. On the basis of the dimensional measurements, the most stable alloys appear to be:

Uranium-3.5 w/o molybdenum-2.0 w/o aluminum
 Uranium-3.5 w/o molybdenum-0.1 w/o silicon
 Uranium-3.5 w/o molybdenum-0.5 w/o silicon

Uranium-7.5 w/o molybdenum-0.1 w/o aluminum
 Uranium-7.5 w/o molybdenum-0.5 w/o silicon

Uranium-10 w/o molybdenum

Relatively large dimensional changes were experienced by specimens with the following compositions:

Uranium-3.5 w/o molybdenum
 Uranium-3.5 w/o molybdenum-0.1 w/o aluminum
 Uranium-3.5 w/o molybdenum-0.5 w/o aluminum
 Uranium-3.5 w/o molybdenum-2.0 w/o silicon
 Uranium-3.5 w/o molybdenum-0.1 aluminum-0.5 w/o silicon

Uranium-5.0 w/o molybdenum

Uranium-7.5 w/o molybdenum
 Uranium-7.5 w/o molybdenum-2.0 w/o niobium

A more meaningful indication of the stability of these alloys will be obtained from the density measurements.

Plans for Future Work

The density of all the specimens will be measured. Metallographic examinations and hardness measurements will be performed on at least 14 specimens. Burnup analyses of selected specimens will be correlated with gamma scans to determine burnup profile.

GENERAL FUEL-ELEMENT DEVELOPMENT (AEC-DRD)

S. J. Paprocki

These studies are being performed for the AEC Division of Reactor Development.

Fabrication of Cermet Fuel Elements

S. J. Paprocki, D. L. Keller, and D. E. Kizer

Objective

To develop fabrication techniques and obtain property data for cermet fuels.

Program Approach

Cermets containing 60 to 80 volume per cent UC are being prepared for physical- and mechanical-property measurements for comparison with values obtained on UN and UO_2 cermets.

Progress Reported Previously

Mechanical-property data for molybdenum-UN and chromium-UN cermets were reported in BMI-1577. Thermal-conductivity values obtained on an 80 volume per cent UC-molybdenum cermet were reported last month in BMI-1581 between 300 and 1100 C. Values ranged from 0.132 to 0.228 w/(cm)(C).

Work This Period

Thermal-conductivity tests have been completed on a 60 volume per cent UC-molybdenum cermet having a density 97 per cent of theoretical. Measured values at 200, 400, 600, 800, 1000, and 1200 C were, respectively, 0.27, 0.29, 0.31, 0.35, 0.38, and 0.41 w/(cm)(C). In addition, mechanical-property measurements were completed on UC-molybdenum cermets. Values are reported in Table D-1. Values are greater than values obtained on the reference UO_2 -molybdenum cermets reported in BMI-1487.

Plans for Future Work

Future studies on cermet fuels will be concerned with more detailed development of molybdenum-base systems.

TABLE D-1. COMPRESSIVE MECHANICAL-PROPERTY VALUES
MEASURED ON UC-MOLYBDENUM CERMETS

Composition, volume per cent UC	Density, per cent of theoretical	Ultimate Compressive Strength, 10^3 psi	Youngs Modulus, 10^6 psi	Yield Strength, 10^3 psi	
				0.1 Per Cent Offset	0.2 Per Cent Offset
60	95	139.5	28.8	114.5	113.8
60	98	111.7	25.5	73.9	80.0
80	95	95.1	27.8	93.5	Not obtained

Development of Low-Cost Tubular Fuel Elements

S. J. Paprocki, R. J. Carlson, and E. G. Smith, Jr.

Objective

To develop techniques for fabricating low-cost tubular fuel elements of consistent quality.

Program Approach

Several techniques for fabricating tubular fuel elements have been considered in this program. One approach has been selected for further development as it appears to offer the best possibility of meeting the basic requirements of low relative cost and consistent quality. The process selected involves the assembly of concentric cladding tubes to form an annulus. The annulus is then loaded with dry-blended fuel and matrix powders, and the tube assembly is sealed. Compaction and sizing of this assembly are accomplished through explosive forming in a die. Sintering of the compacted annulus and core-to-cladding bonding are achieved by gas-pressure bonding.

Another closely related implosive-compaction technique has shown considerable promise in the limited tests made to date. Assembly and loading are the same as for the explosive technique while a mandrel and outside sleeve are employed to jacket the concentric fuel tubes and core. Explosive sheet is wrapped around the outside of the assembly and detonation takes place under water.

Progress Reported Previously

Studies to date for the concentric-component approach have resulted in a reference procedure being established for loading, compacting, and bonding tubular fuel elements. Loose-powder loading of the annulus formed by the concentric cladding tubes while heating the entire assembly results in the most uniform and highest as-loaded densities. Compaction and sizing of the as-loaded fuel tubes are accomplished by the use of explosives in a special two-part die assembly. The as-compacted tubes exhibit rather good dimensional control. While the inside surfaces are somewhat wrinkled, except for some dimples produced, the outside surfaces are smooth. The dimpling of the tubes is attributed to areas of initial low density and is accordingly considered a loading problem. Metallographic examination of the core indicates a near theoretical density, but the UO_2 dispersion is not uniform, and this reflects the blending operation, which at present consists only of dry blending of the fuel powders.

Tubes compacted by the implosive technique exhibited a core density near theoretical as determined by metallography. In addition, some areas of core-cladding bonding were noted in the as-compacted tubes. The original spherical UO_2 particle shape was observed to have been flattened out by the action of the explosive.

Work This Period

Metallographic examination of Tubes 38, 40, and 41 has been completed. Tube 38 was pressure bonded at 2100 F and 10,000 psi for 3 hr and the core exhibited a near-theoretical density. The core-cladding bonding was observed to be complete for both the longitudinal and transverse sections. Nonuniformity of core thickness and lack of a uniform dispersion were noted and attributed to inadequate loading and blending processes. Tubes 40 and 41 were hydrostatically pressed at 10,000 and 50,000 psi, respectively, prior to being implosively compacted using a charge density of 2.5 g per in.². Some degree of core-cladding bonding was noted both for specimens as well as areas of separations. A near-theoretical core density was observed for both tubes. The special blending technique employed for these tubes was partially effective in improving the uniformity of dispersion and, together with the hydrostatic predensification

step, eliminated some of the distortion of the spherical UO_2 particles during the implosive-compaction operation.

Plans for Future Work

The experimental portion of this program is now completed, and a topical report will be prepared.

Gas-Pressure Bonding of Ceramic Fuel Elements

S. J. Paprocki, S. W. Porembka, E. S. Hodge, and C. B. Boyer

To develop and evaluate the gas-pressure-bonding process as applied to vibratory-compacted fuel rods supplied by GE-Hanford and rolled UO_2 sheet materials as supplied by Gladding, McBean & Company.

Program Approach

The present program concerns two areas of research which represent cooperative programs with Hanford and Gladding, McBean & Company. In these programs, the gas-pressure-bonding process as combined with vibration powder-packing techniques and UO_2 sheet-rolling techniques is being investigated. The vibration packing-pressure bonding studies are directed toward the definition of practical vibration-packed UO_2 materials which will provide the required initial density and ceramic activity for successful pressure bonding. Flat-plate designs consisting of pressure-bonded sheet-rolled UO_2 will serve to demonstrate unique low-cost fuel-element designs.

Progress Reported Previously

In previous studies, the evaluation of various types of uranium dioxide powders has been carried out on the basis of their initial compacting and pressure-bonding characteristics. The results of this work and the development of pressure-bonding techniques for various fuel geometries were discussed in two phase reports, BMI-1475 and BMI-1555. Four Type 304 stainless steel-clad UO fuel rods fabricated by pressure bonding are presently being irradiated in the VBWR.

Initial tests with pressure-bonded sheet-rolled UO_2 platelets have demonstrated behavior similar to that expected from pressed and sintered cores. A slight tendency toward cracking during bonding has been reduced by altering the rate of pressure application during heating. Cladding of these platelets with both stainless steel and Zircaloy-2 has been demonstrated.

Five groups of vibration-packed UO_2 fuel rods as clad with stainless steel and Zircaloy-2 have been pressure bonded. Major emphasis has been placed in the areas of optimizing particle sizing of powder components, evaluating oxide mixtures containing Micronized UO_2 , and minimizing cladding thickness. Intermediate swaging has also been considered in these tests.

Work This Period

The sixth series of stainless steel-clad UO_2 rods received from Hanford last month was gas-pressure bonded by first pressurizing to 6,000 psi prior to heating, followed by increasing temperature and pressure to the operating conditions of 2100 F at 10,000 psi. These bonding conditions were maintained for 3 hr.

Of the 13 rods described in Table D-2 seven failed prior to or during bonding because of cracking of the cladding near the end cap or collapsing of the hollow end caps used in several of these elements. These problems are being corrected on future rods. Two of the six rods that were successfully pressure bonded, 68-2 and 74-1, were

TABLE D-2. COMPOSITION AND INITIAL DENSITIES OF VIBRATION-PACKED AND SWAGED STAINLESS STEEL-CLAD UO_2 RODS IN THE SIXTH SERIES

Rod	Oxide Composition			Fabrication Method	Cladding Thickness, in.	Density ^(a) , per cent of theoretical
	Amount, w/o	Mesh Size	Powder Type			
63-1 and 63-2	70 30	-200	Fused Micronized	Vibration packed and cold swaged from 0.750-in. OD to 0.574-in. OD	.010	78.4/77.7
66-1 and 66-2	70 30	-100 + 200	Fused Micronized	Vibration packed and cold and hot swaged from 0.750-in. OD to 0.573-in. OD	.010	84.8/83.3
68-1 and 68-2	70 30	-65 + 100	Fused Micronized	Vibration packed and cold and hot swaged from 0.750-in. OD to 0.573-in. OD	.010	84.6/77.9
69-1 and 69-2	70 30	-35 + 65	Fused Micronized	Vibration packed and cold swaged from 0.750-in. OD to 0.575-in. OD	.010	81.0/79.0
73-1 and 73-2	70 30	-20	Fused Micronized	Vibration packed and cold swaged from 0.750-in. OD to 0.575-in. OD	.010	82.0/81.7
74-1 and 74-2	70 30	-20	Fused Micronized	Vibration packed and cold and hot swaged from 0.750-in. OD to 0.570-in. OD	.010	84.0/82.0
4	55	-4 + 8	Fused	Vibration packed and cold swaged one pass from 0.563-in. OD to 0.538-in. OD	.009	89.0
	12.5	-10 + 20	Fused			
	12.5	-35 + 65	Fused			
	20	-200	Fused			

(a) Swaged densities.

returned to Hanford intact. Final density and stoichiometry measurements were made at various points along the length on the four rods retained for evaluation at Battelle. The data obtained in these tests are listed in Table D-3.

TABLE D-3. FINAL DENSITY AND STOICHIOMETRY MEASUREMENTS ON THE SIXTH SERIES OF GAS-PRESSURE BONDED STAINLESS STEEL-CLAD UO_2 RODS^(a)

Rod	Average Density After Pressure Bonding, per cent of theoretical	Oxygen/Uranium Ratio After Pressure Bonding
66-2	95.6	2.002
	95.3	2.010
	94.4	2.006
68-1	95.4	2.012
	94.4	2.016
	92.7	2.004
69-1	95.5	2.006
	95.3	2.006
	94.4	2.004
74-2	95.6	2.014
	96.3	2.012
	95.4	2.000

(a) Samples taken at three locations down the length of the rod.

In general, these six rods had smooth cladding surfaces and remained fairly straight. They also appeared to have been very uniformly packed. Metallographic examinations are being performed to establish the uniformity of distribution for these oxide mixtures.

Plans for Future Work

Further vibration-packed and swaged stainless steel-clad UO_2 rod specimens are planned. These specimens will incorporate finer particle sizes and greater additions of the Micronized UO_2 . An interim progress report on this program is being initiated. A final report on the pressure bonding of stainless steel-clad sheet-rolled UO_2 is also being written.

Factors Affecting Pressure Bonding

D. C. Carmichael and J. W. Spretnak

Objective

To determine the mechanism of solid-phase bonding of metal components under the application of heat and pressure.

Program Approach

Hot pressing or gas-pressure bonding under various conditions of temperature, pressure, and time is being used to prepare specimens for study. The specimens are of different designs and preparations and are composed of two components of the same

or different metals. Currently, couples consisting of two platelets of the same metal which are 1/2 in. square and 1/8 in. thick are being vacuum hot pressed in limited number. Evaluation of the bonds is based chiefly on metallographic examination using special techniques, as described in BMI-1512. Observations are made of the extent of contact between mating surfaces and of the behavior of grain boundaries, dislocations, and interfacial voids which affect the elimination of the interface during bonding.

Progress Reported Previously

The mechanism of the self-bonding of copper has been established, and the effects of pressure, temperature, grain growth, impurities, and surface roughness have been investigated. It was found that the first basic requirement for producing a metallurgically perfect bond is the application of sufficient pressure to place the surfaces in intimate contact, which for copper could be related to Meyer hot-hardness values. The second requirement was found to be the minimization of voids in the bond interface, which prevent grain growth across the interface. These results are discussed in detail in a topical report, BMI-1512.

The pressure-bonding mechanism is now being studied in more detail and in other metals than copper. Results of recent studies of the deformation behavior during formation of bonds in nickel-copper couples as compared to copper-copper couples were discussed in a previous progress report, BMI-1549. Results of an investigation of this phase of the bonding process for aluminum self-bonding were reported principally in BMI-1561 and BMI-1565. A study of the effect of recrystallization during bonding on the elimination of the bond interface was initiated using nickel couples.

Work This Period

The investigation of the role of recrystallization in the bonding mechanism has been continued. The effects of bulk recrystallization of cold-worked bonding components and of localized recrystallization of components with worked surfaces are being studied. The specimens presently being used consist of cold-rolled pure nickel platelets; a rough, shaper-finished surface is being bonded to a smooth, ground surface. Microhardness and metallographic examinations have been completed on couples vacuum-hot pressed at 1000 and 1400 F and at 10,500 and 4,700 psi, respectively. Partial recrystallization of the cold-worked components occurred during the bonding cycle at 1000 F. The bulk hardness of the platelets was reduced from about 322 to 167 Knoop. Bonding at 1400 F resulted in complete recrystallization and some grain growth. The hardness was reduced to about 147 Knoop. However, the interface in both cases showed a continuous thin film of contamination, and the grain growth was not forced across the interface by recrystallization of either the bulk structure or the cold-worked, shaper-finished surface. No significant difference in the microhardness of the shaper-worked surfaces as compared to the cold-worked bulk material could be shown prior to bonding; therefore, the effect of the surface could not be established in these cold-worked specimens.

Plans for Future Work

Additional samples will be bonded to continue the investigation of the effects of localized and bulk recrystallization on pressure bonding. The components will be in different annealed and cold-worked states and will be bonded using various temperatures and pressures. Initial emphasis will be placed on determining techniques for eliminating or minimizing the contamination film in the bond interface.

PREPARATION OF UO_2 SINGLE CRYSTALS (AEC-DR)

F. A. Rough

Two studies for the AEC Division on the preparation of single-crystal UO_2 are reported in this section.

Growth of UO_2 Single Crystals

J. D. Jackson, C. A. Alexander, and H. L. Goering

Objective

To prepare large single crystals of UO_2 .

Program Approach

This program is being supported by the AEC Division of Research. An apparatus to allow crystal growth from the melt by the Bridgman technique is being developed.

Progress Reported Previously

Several high-temperature sources have been investigated. Effort was expended in development of a tungsten-resistance furnace which is capable of reaching the 2800 C temperature necessary to melt UO_2 . Experiments have shown that elemental uranium occurs in the UO_2 boule when operating in argon or vacuum (as required for experiments in this furnace) and considerable porosity is encountered. During the remaining program, the growth of UO_2 crystals will be investigated by the Verneuil technique, using as a heat source an induction-coupled plasma in which oxidizing, reducing, and neutral atmospheres can be investigated.

Work This Period

During June, experimentation with the induction-coupled plasma was concluded. The equipment was modified by using a single, 25-mm-ID, quartz tube to contain the plasma. Closer coupling of the plasma in this tube made possible attainment of higher temperatures on larger boules. For instance, a 3/8-in. -diameter sintered UO_2 rod could be melted and a 1/8-in. -thick molten pool maintained by the plasma.

UO_2 powder (through 100 mesh) was fed onto the molten boule in several experiments. By control of the gas flow and the distance of the feed tube above the plasma, most of the UO_2 powder could be made to impinge directly on the boule. This was shown by the blue coloration given to the plasma by the feed.

In these experiments, the boule was not seeded, nor was it grown to very small size to form a single crystal. However, some crystal growth was obtained, and crystals as large as about 1/16 in. were seen on the surface of the fused boule.

Metallographic examination of two fused boules was made. A definite fused area about 1/16 in. thick was seen above the sintered rod. No cracking was observed in the fused area even though the sample cooled rapidly when the plasma was turned off. Some cracking was found in the sintered rods on heating, but was usually observed before the entire sample reached red heat.

Examination of the microstructure revealed considerable amounts of free uranium, up to several per cent. (In a previous experiment, with oxygen in the feed gas, considerable oxidation was obtained.) The free uranium was found both along what appeared to be grain boundaries, and uniformly throughout the crystal. In addition, some pores of about the same size and distribution as the uranium inclusions were seen. (These could be simply pockets from which inclusions were detached during polishing.)

Plans for Future Work

No further study is planned. Results obtained in the limited work on the plasma Verneuil suggest the method will be applicable to the growth of UO_2 crystals, if sufficient control of oxygen content of the gas is provided.

Characteristics of Heat-Treated UO_2 Crystals

M. J. Klein, A. A. Bauer, and F. A. Rough

Objective

To establish a reliable heat-treating procedure to produce single-phase stoichiometric UO_2 crystals from production-grade material in sizes suitable for physical-property measurements.

Program Approach

Fused UO_2 crystals obtained from three different suppliers were examined and analyzed to establish their stoichiometry, purity, and homogeneity. Impurities and second-phase inclusions were identified where possible, and the relative quality of UO_2 crystals was established for each supplier.

The optimum lots of UO_2 crystals were selected, and a series of heat-treating tests, designed to improve the stoichiometry and decrease the second-phase impurities, was conducted.

Upon conclusion of the heat-treating tests the quality of these crystals was again re-established and compared to the quality of the crystals as received from the suppliers.

Progress Reported Previously

Crystals from one supplier were judged to be too small to be suitable for physical-property measurements. Consequently, studies were continued only with Norton and Spencer crystals.

Spectrographic analyses of Norton and Spencer crystals showed them to be of good chemical purity, having less than 50 ppm total detectable metallic impurities. U_2N_3 and uranium were detected as impurity phases in Norton crystals. Crystals from both suppliers were heat treated for 1 hr at 1900 C in a vacuum and in a hydrogen atmosphere. It was found that an impurity phase was still present after the hydrogen heat treatment. The vacuum-treated crystals were found to contain scattered porosity throughout the matrix, while none of the impurity phases originally present in these crystals could be detected.

Work This Period

Crystals were heat treated in an argon atmosphere for 1 hr at 1900 C and in a hydrogen atmosphere for 10 hr at 1700 C. Impurity phases were still present in the crystals after these treatments.

Laue X-ray photographs were obtained of the as-received crystals and of the crystals after each heat treatment to determine the relative degree of crystal perfection. It was found that vacuum heat treatments introduced a substructure into the crystal matrix which was also visible metallographically. Crystals heat treated in hydrogen at 1900 C also evidenced a decrease in crystal perfection. Argon-treated crystals and crystals heated in hydrogen at 1700 C showed crystal perfection of about the same degree as the as-received crystals.

Plans for Future Work

No further work is planned.

RADIOISOTOPE AND RADIATION APPLICATIONS (AEC-DID)

D. N. Sunderman

These studies are in support of the program of the AEC Division of Isotopes Development, Isotopes Technology and Production and Radiation Branches.

Use of Intrinsic Radioactive Tracers for Process Control

P. Gluck, J. F. Kircher, C. W. Townley, and D. N. Sunderman

Objective

To investigate the feasibility of using in-process isotope neutron sources to produce short-lived intrinsic radiotracers useful for steam analysis and process control.

Program Approach

The approach being used is to determine first the optimum practical conditions for in-process activation of steam constituents to form short-lived intrinsic radiotracers. This work is being accomplished by activating standard solutions of salts such as manganese sulfate, as well as indium foils, while varying such parameters as activation volume, geometry, holding time, etc. Other experiments are being performed to determine general steam activation efficiencies for different process steams as a function of neutron-source strength. Finally, the concept of in-process neutron activation will be tested through the production of very short-lived isotopes which should be of utility in process control under plant operating conditions.

Progress Reported Previously

The present phase of the intrinsic radiotracer program began July 1, 1961. After an initial survey of neutron sources and source technology, 10-curie polonium-beryllium source was obtained from Mound Laboratory. This source was used for preliminary work. A combination neutron-source storage shield and activation cell was constructed for use in the experimental program.

Modification of the activation cell into a flow system and installation of the 50-curie polonium-beryllium source were reported last month in BMI-1581. This completed preparation for the final phase of the program. Results were reported for the neutron and gamma radiation levels in the area. The initial data for activation of manganese and indium solutions in the flow system were also reported.

Work This Period

Activation experiments with the flow system were continued this month. Solutions of vanadium oxide in dilute nitric acid and silver nitrate in ammonium hydroxide and in ammonium thiosulfate plus ammonium hydroxide were activated at flow rates ranging from 50 to 595 ml per min. Nitric acid solutions of silver were also used, but difficulties were encountered with silver plating out on the stainless steel; so these solutions were discontinued. The solution activity levels were measured in both the 750-cm³ dip counter with the 2 x 2 NaI(Tl) crystal and the 340-ml beta-counting cell with the seven Anton No. 112 probes.

Table H-1 gives the equilibrium silver and vanadium activities in counts per minute per gram of metal per liter. The results are normalized to a 50-curie polonium-beryllium source. Saturation activities were calculated from an activation-cell residence

TABLE H-1. EQUILIBRIUM ACTIVITIES IN VANADIUM AND SILVER SOLUTIONS, DUE TO
A 50-CURIE POLONIUM-BERYLLIUM SOURCE, AS A FUNCTION OF SOLUTION
THROUGHPUT

Concentration of Metals in Solution, g per liter	Flow Rate, ml per min	Activation Time, min	Activity, cpm per g of metal	
			Equilibrium	Saturation
<u>Gamma Activity</u>				
0.2 vanadium	50	48	15,020	15,020
	100	24	20,050	20,070
	200	12	30,970	34,800
	300	8	33,700	43,700
	400	6	31,700	47,400
	500	4.8	29,550	50,300
0.2 silver ^(a)	200	12	2,755	2,755
	300	8	4,055	4,055
	400	6	4,550	4,550
	500	4.8	5,870	5,870
	595	4	7,240	7,240
0.5 silver ^(b)	100	24	945	945
	200	12	2,435	2,435
	300	8	3,660	3,660
	400	6	4,670	4,670
	500	4.8	5,950	5,950
	595	4	7,560	7,560
<u>Beta Activity</u>				
0.5 silver ^(b)	100	24	2,900	2,900
	200	12	7,180	7,180
	300	8	10,580	10,580
	400	6	12,760	12,760
	500	4.8	16,400	16,400
	595	4	19,850	19,850
0.2 vanadium	50	48	4,910	4,910
	100	24	7,240	7,330
	200	12	8,910	10,010
	300	8	9,435	12,220
	400	6	8,900	13,300
	500	4.8	8,220	14,000
	595	4	7,660	14,600

(a) Silver complexed with ammonium hydroxide.

(b) Silver complexed with ammonium thiosulfate and ammonium hydroxide.

time based on the flow rate. The activation products assumed in each case were vanadium-52 and silver-110 with half-lives of 3.76 min and 24 sec, respectively. In all cases the saturation activity was found to increase with flow rate. This is apparently due to decay of the short-lived activation products as the solution flows through the counting cells.

Plans for Future Work

Additional activations will be performed next month. A comprehensive re-evaluation of the information obtained thus far will be initiated so that it may be presented in the form most useful for engineering applications.

Graft-Polymerization Studies

F. A. Sliemers, M. Luttinger, R. W. Godwin, W. B. Gager,
R. Lieberman, J. F. Kircher, and R. I. Leininger

Objective

To determine the influence of structural and compositional factors on the behavior of polymers subjected to gamma irradiation and to correlate the radiation-induced changes which occur with the ability of the polymer to form graft copolymers.

Program Approach

It is anticipated that a mechanism of radiation attack on the various polymer systems can be established. Polymers are irradiated in vacuo to various total doses. Free-radical site concentrations are determined both as a function of dose and as a function of time, using electron paramagnetic resonance (EPR) measurements. Vapor-phase chromatography and mass spectrometry are used to determine quantitatively the volatile fragments of the irradiation. Grafting techniques are employed to correlate free-radical site concentrations and grafted copolymer.

Progress Reported Previously

A number of acrylate and methacrylate polymers have been examined extensively. The data suggest that (1) free-radical formation is generally accomplished by means of an ester scission, (2) ester scission is accompanied, in most cases, by a scission on the polymer backbone, (3) the size and configuration of the hydrocarbon tail of the ester group and of the group attached to the carbon atom alpha with respect to the ester group influence the efficiency of site formation, and (4) the rate of formation and/or decay of free-radical sites is affected by polymer mobility and, perhaps independently, by polymer molecular weight.

Work This Period

During June, experimental work included (1) extension of the peroxide-initiated grafting studies to the system polymethylmethacrylate-vinyl pyrrolidone, (2) determination of site concentration as a function of dose in polymethyl- α -bromoacrylate, and (3) additional polymer preparation work.

The peroxide-initiated grafting work during the month included the system polymethylmethacrylate-vinyl pyrrolidone. The absence of detectable amounts of grafting to polymethylmethacrylate in several postirradiation studies completed earlier in the program has been attributed to solubilization effects. Apparently, during solubilization, free-radical decay proceeds at a much faster rate than does the propagation step of the copolymerization reaction. The decay is probably due to the large increase in molecular mobility which occurs as solubilization proceeds.

In the present study, both undiluted monomer and a 50 per cent solution of the monomer in water were reacted with the irradiated peroxidized polymer at 120 C. The diluent was used in order to reduce the solubility of the polymer in the monomer. Grafting was carried out at three different radiation doses. The polymer samples are now being analyzed for nitrogen.

In addition to the peroxide-initiated grafting studies, an attempt was made to graft acrylonitrile to irradiated polymethylmethacrylate using vapor-phase grafting techniques. Again, the object of the work has been to reduce the mobility of the polymer molecules during the copolymerization reaction. In this work, dry nitrogen was used as a carrier gas for the monomer vapor. Approximately 100 ml of acrylonitrile was passed through the system during a 7-hr run at ambient temperatures. A nitrogen analysis is now being performed to establish the extent of grafting.

Samples of polymethyl- α -bromoacrylate, irradiated to various dosages, were subjected to EPR measurements to determine the sites-versus-dose relationship. Interestingly, no measurable numbers of sites were formed even at a total dose of approximately 3.4×10^7 rads. The molecular weight of this material will be determined before it is compared, in terms of radiation susceptibility, with other α -substituted acrylate polymers.

In the polymer-preparation phase of the program methyl- α -bromoacrylate polymer was synthesized. Also, work was directed toward the synthesis of the α -phenyl and α -iodo derivatives of polymethylacrylate.

Plans for Future Work

Work during July will be concentrated in the several areas described above.

A Study of the Effect of Internal Irradiation on Catalyst Activity

J. L. McFarling, J. F. Kircher, and D. N. Sunderman

Objective

To investigate the feasibility of obtaining enhanced catalytic activity by internal irradiation of catalysts.

Program Approach

The planned approach for this program is as follows: (1) catalysts will be prepared containing a small percentage of ceramic-coated UO_2 particles dispersed throughout the matrix, (2) the catalyst mixtures will then be irradiated in the Battelle Research Reactor to produce fission-product activity in the fuel particles, and (3) the radioactive catalysts will be compared with unirradiated catalysts with regard to their catalytic activity.

Progress Reported Previously

The reaction being studied is the catalytic dehydration of cyclohexanol to cyclohexene at 400 C over a magnesium sulfate-calcium chloride catalyst. The vapor-deposition group at Battelle has supplied the coated-fuel particles for the study. A simple glass-tube reactor fitting into a small tube furnace is being used for the experimental study. Conversion efficiency of the catalyst is being determined by gas chromatographic analysis of the product.

Work This Period

Further experiments using unirradiated catalysts have indicated the desirability of changing the catalyst composition somewhat. The catalyst composition now being studied is the following:

	Amount, w/o	
	Unfueled Catalyst	Fueled Catalyst
MgSO ₄	35	33.25
CaCl ₂	65	61.75
UO ₂ fuel particles	--	5

This catalyst composition has given quite reproducible conversion efficiencies, as shown in Table H-2. The fuel particles used in these experiments were about 100 μ in diameter with a 20- μ pyrolytic-graphite coating. The vapor-deposition group has also supplied 100- μ fuel particles coated with aluminum oxide.

TABLE H-2. DEHYDRATION OF CYCLOHEXANOL USING FUELED AND UNFUELED CATALYST

Run	Description of Catalyst					Alcohol Throughput Rate	Temperature, C	Conversion After 250 Min, per cent
	Composition, w/o			Physical Form	Weight, g			
	MgSO ₄	CaCl ₂	UO ₂ Particles					
27	35	65	--	Granulated	0.25	0.20 $\pm$ 0.01	400	50
28	35	65	--	Granulated	0.25	0.20 $\pm$ 0.01	400	5.7
29	35	65	--	Granulated	0.25	0.20 $\pm$ 0.01	400	5.3
31	33.25	61.75	5	Granulated	0.25	0.20 $\pm$ 0.01	400	5.2
32	33.25	61.75	5	Granulated	0.25	0.20 $\pm$ 0.01	400	6.1

A sample of fuel particles was irradiated for a short time in the Battelle Research Reactor and beta and gamma activities are being followed as a function of fission-product decay time.

Plans for Future Work

The experimental runs using reactor-irradiated catalyst mixtures will begin as soon as the samples have been irradiated. Tests on both fueled and unfueled irradiated catalyst mixtures are planned for next month.

Establishment of Test Parameters for Sealed Radiation Sources

H. S. Rosenberg, R. A. Ewing, and D. N. Sunderman

Objective

To identify and evaluate the stresses to which sealed radiation sources are exposed in their major areas of application, to analyze the major parameters involved in testing sources for their resistance to normal environmental conditions, and to recommend procedures for accelerated testing.

Program Approach

Sufficient data are to be accumulated to establish the ranges of environmental conditions to which sealed sources may be exposed in each major area of application. From these data recommendations can be made for accelerated test procedures to evaluate resistance to normal environmental conditions. Also, test parameters for selected major abnormal environments will be recommended. Some of the necessary information will be obtained from applications and licenses for by-product materials. To complete the information, selected manufacturers of sources and devices incorporating sealed sources as well as users of sources will be contacted.

Progress Reported Previously

Work on this program has just begun and no previous reports have been made.

Work This Period

During the month arrangements were made with the Division of Licensing and Regulation of the AEC to inspect the file of applications and by-product licenses. To avoid inspection of the over 8000 licenses on file, a copy of AEC List "D" was obtained and a preliminary screening of these licensees is being made. To date, approximately 250 licensees, principally in the Midwest, have been further categorized by industry and by location.

Plans for Future Work

Next month it is expected that the list of license applications to be examined will be completed and the actual survey of the files initiated.

MATERIALS DEVELOPMENT AND EVALUATION (HAPO)

F. R. Shober

Research and development work being conducted on materials to be used in conjunction with construction of the NPR (New Production Reactor) and operation of the PRTR (Plutonium Recycle Test Reactor) at Hanford Atomic Products is reported in this section.

Mechanical Properties of Zirconium Alloys

J. A. Van Echo

Objective

To obtain creep and creep-rupture data, especially long-term creep, on annealed and cold-worked Zircaloy-2 sheet at 550, 650, and 750 F and to evaluate the effect of stress and/or temperature cycle on the creep properties of this alloy.

Program Approach

The tests are being conducted in standard Battelle creep machines and furnaces. In order to prevent excessive contamination of the specimens, all tests are made in vacuum of the order of 0.5 to 0.1 μ . Creep measurements are taken approximately daily by means of an optical system. In all respects, the tests meet or exceed the requirements for creep testing specified in ASTM Standard E-139-58T.

In the stress- and temperature-cycle tests, the stress is removed first and then the temperature is lowered to room temperature 1 day per week. The stress is re-applied after the specimen has been returned to test temperature. No temperature-cycle tests are being conducted at present but these will be resumed in the near future.

Progress Reported Previously

All twenty tests now in progress have been in progress for times ranging from about 5500 to 40,700 hr. Data have been accumulating for some time and have been reported in progress reports from time to time. The previous summary was presented in the May progress report (BMI-1581). Since all tests are long-term, there is little change in test results, other than accumulated time and small increases in total creep strain, from one month to the next. In some cases, the creep rate changes, but this is generally very slight. Relative changes in creep strain can be observed by comparing the data given in the February (BMI-1569) and May (BMI-1581) reports.

Work This Period

All 20 tests reported in progress as of the end of May were continued through this period. These tests consist of the following:

- (1) Two creep tests on 15 per cent cold-worked material at 650 and 750 F with test times of about 38,300 and 40,700 hr
- (2) Nine creep tests on annealed sheet at 550, 650, and 750 F with test times ranging from about 21,600 to 28,700 hr
- (3) Five temperature- and stress-cycled tests on annealed material at 550 and 650 F; with test times ranging from 14,300 to 17,800 hr
- (4) Four creep tests on annealed PRTR pressure-tube material at 450 and 550 F with test times ranging from 5,600 to 13,200 hr.

All tests continued at approximately their established creep rates. No additional tests have been started and none discontinued during this period.

Plans for Future Work

The 20 tests in progress are scheduled to continue. Creep data will be accumulated until a failure has occurred or it is believed that continuing a test will no longer contribute useful data. As tests are discontinued, new tests will be started on specimens taken from the PRTR or the NPR pressure tubes. In addition, more tests are scheduled to be made on specimens which are stress and/or temperature cycled.

Thermal Conductivity and Electrical Conductivity of UO_2

H. W. Deem and J. Matolich, Jr.

Objective

To measure the thermal and electrical conductivity of irradiated and unirradiated UO_2 .

Program Approach

Existing apparatus with some modifications will be used to make these measurements. The apparatus and modifications were described in BMI-1569. Specimens were also described in BMI-1569.

Progress Reported Previously

The longitudinal specimen apparatus was modified and reassembled. The first set of thermal-conductivity measurements were concluded on the fused single-crystal UO_2 specimen. Tentative data were reported in BMI-1581.

Work continued on the hot-cell apparatus modification.

Thermal-conductivity measurements on the disk-shaped UO_2 specimen were suspended pending receipt of additional data from HAPO.

Work This Period

Thermal-conductivity measurements have been concluded on the fused single-crystal UO_2 specimen after its first irradiation at HAPO. Tentative values are given in Table J-1. Specimen irradiation exposure is not known at this time.

TABLE J-1. TENTATIVE INTERPOLATED THERMAL
CONDUCTIVITY OF FUSED SINGLE-
CRYSTAL UO_2 AFTER RADIATION
EXPOSURE 1

Temperature, C	Thermal Conductivity, w/(cm) (C)
200	0.058
400	0.045
600	0.040
800	0.040
900	0.040

Plans for Future Work

During the next work period the third set of measurements will be made on the fused single-crystal UO_2 specimen which will have received a second radiation exposure at HAPO. The specimen then will be returned to HAPO for further irradiation.

Final assembly is being made on the hot-cell apparatus. After assembly measurements will be made on Specimen 51, and on the new set of irradiated specimens.

COATED-PARTICLE FUEL MATERIALS (AEC-DRD)

R. W. Dayton, J. H. Oxley, and C. W. Townley

This program is being carried out for the AEC Division of Reactor Development to determine the characteristics of various coated-particle fuels and to develop optimum materials for specific reactor applications.

MATERIALS DEVELOPMENT PROGRAMUncoated Fuel-Particle Studies

G. W. Cunningham, J. M. Fackelmann, and S. J. Paprocki

Objective

To characterize and evaluate fuel particles of UO_2 , UC, and UC_2 for use in coating studies and in subsequent coated-fuel irradiation programs.

Program Approach

Fuel materials are purchased as required from commercial vendors and are characterized with respect to enrichment, composition, particle size, porosity, shape factor (sphericity, roundness, and surface roughness), hardness, crushing strength, phase structure, grain size, fraction of particle agglomerates, surface-activity ratio, xenon-release ratio, and chemical impurities. As the relative importance of the various factors becomes known, emphasis will be increased or decreased on these factors.

Progress Reported Previously

Various batches of enriched UO_2 and UC_2 powder have been characterized with respect to the factors described above. Results are tabulated in Tables L-1 and L-2.

Work This Period

No new material has been received for evaluation.

Plans for Future Work

Lots of UC, UO_2 , and UC_2 will be evaluated as received.

Alumina-Coating Studies

M. F. Browning, V. M. Secrest, and J. M. Blocher, Jr.

Objective

To establish conditions and equipment design for coating fuel particles with Al_2O_3 of optimum quality.

Program Approach

Alumina is being deposited on UO_2 particles by hydrolysis of Al_2Cl_6 vapor in a fluidized bed. The coatings are being evaluated by microscopic examination, measurement of physical properties, alpha counting, and measurement of fission-gas release on postirradiation heating. Promising materials will be evaluated by in-pile tests at elevated temperatures.

TABLE L-1. SUMMARY OF DATA FOR UNCOATED UO_2 PARTICLE CHARACTERIZATION

	Batch UO_2 -E1	Batch UO_2 -E2	Batch UO_2 -E3	Batch UO_2 -E4	Batch UO_2 -E5	Batch UO_2 -E6	Batch UO_2 -E7	Batch UO_2 -E8
Enrichment, per cent	92.0	92.0	93.18	93.0	92.7	92.9	93.0	93.0
Oxygen/Uranium Ratio	2.012	2.004	2.004	2.039	2.054	2.003	2.002	2.002
Average Diameter, μ	112	112	122	122	122	122	178	122
Density, per cent of theoretical	~80	98	97.8	83	87	92	92	93
Spheroidal Particles, per cent	~95	~95	95	15	17	92	94	95
Relative Surface Area	40.8	--	2.67	15.5	17.8	6.67	13.00	4.45
UO_2 Phase, volume per cent	88	93	87.8	85.2	70.2	75.7	85.4	84.4
Particle Agglomeration, per cent	--	--	0.2	5.0	2.0	2.0	1.0	1.0
Grain Size, μ								
Average	--	15	8.8	4.2	3.0	5.3	5.5	4.8
Maximum	--	50.4	43.8	25.9	11.6	40.5	24	22.2
Impurities, ppm								
Aluminum	--	--	--	100	100	100	200	200
Boron	<1	<0.05	<1	--	--	--	0.5	0.5
Cadmium	--	--	--	--	--	--	0.2	0.2
Calcium	30	7	--	15	50	15	20	40
Chromium	20	<3	10	10	10	40	5	5
Copper	2	2	5	2	2	2	4	10
Iron	10	<5	10	100	100	80	10	10
Lead	--	--	--	--	1	25	5	5
Magnesium	2	1.5	3	400	25	3	2	2
Manganese	--	--	<1	1	5	40	1	1
Molybdenum	30	20	20	2	2	2	30	30
Nickel	2	<2	1	3	3	80	4	4
Silicon	30	7	10	200	200	--	10	15
Tin	--	--	<1	--	--	--	--	2
Vanadium	--	--	--	--	--	--	5	5
Cobalt	--	--	--	--	--	--	1	1

TABLE L-2. ANALYTICAL DATA FOR UNCOATED UC₂ PARTICLES

	Batch UC ₂ -E1	Batch EC ₂ -E2	Batch UC ₂ -E3	Batch UC ₂ -E4	Batch UC ₂ -E5
Enrichment, per cent	93	93.2	93	93.4	93.1
Density, g per cm ³	--	11.0	10.8	10.9	10.9
Carbon Content, w/o	--	9.45	9.54	9.21	9.39
Spheroidal Particles, per cent	84	100	100	100	99
Agglomeration, per cent	1	0	0	0	0
Impurities, ppm					
Aluminum	--	10	80	160	200
Boron	--	<1	1	1	1
Beryllium	--	--	0.05	0.1	0.1
Cadmium	--	--	<0.2	<0.2	<0.2
Copper	--	15	5	<1	<1
Cobalt	--	--	5	5	5
Chromium	--	100	80	160	160
Iron	--	300	250	250	250
Magnesium	--	20	5	5	5
Manganese	--	<1	<2	<2	<2
Molybdenum	--	50	120	120	120
Nickel	--	80	160	160	160
Phosphorus	--	--	<40	<40	<40
Lead	--	--	2	2.5	<1
Vanadium	--	5	2	20	<2
Tungsten	--	--	<125	<125	<125
Silicon	--	250	<160	<160	<160
Zinc	--	--	<20	<20	<20
Calcium	--	2	10	10	20
Barium	--	--	<2	<2	<2
Tin	--	--	<1	<1	<1
Titanium	--	20	40	40	40
Arsenic	--	--	<20	<20	<20

Progress Reported Previously

All coatings prepared at 1400 C even though relatively free of impurities exhibit a coating integrity inferior to that of the better 1000 C coatings. Surface protrusions of 1000 C Al_2O_3 coatings can be minimized by interrupting the growth pattern with a thin layer of Al_2O_3 deposited at a lower temperature.

The Al_2O_3 -coated enriched fuel particles listed in the following tabulation have been submitted for in-pile evaluation.

Lot	Fuel Particle		Coating Temperature, C	Coating Description ^(a)	Xenon Release Ratio ^(b) , ppm
	Type	Diameter, μ			
715A	UO ₂ -E2	127	1000	42 μ of dense Al_2O_3	3.2
715A-HT	UO ₂ -E2	127	1000	42 μ of dense Al_2O_3	0.17
717B	UO ₂ -E1	127	1000	43 μ of dense Al_2O_3	0.8
721C	UO ₂ -E3	127	1000	8 μ of dense Al_2O_3	0.98
			700	18 μ of porous Al_2O_3	
			1000	28 μ of dense Al_2O_3	
725D	UO ₂ -E3	127	1225	56 μ of dense Al_2O_3	0.33
732D	UO ₂ -E4	127	1025	43 μ of dense Al_2O_3	0.11
733B	UO ₂ -E6	127	1025	47 μ of dense Al_2O_3	0.03
734C	UO ₂ -E6	127	1300	45 μ of dense Al_2O_3	0.04
735C	UO ₂ -E8	127	1025	16 μ of dense Al_2O_3	0.07
			1250	27 μ of laminar PyC	
736B	UO ₂ -E6	127	1400	14 μ of laminar PyC	0.11
			1025	29 μ of dense Al_2O_3	

(a) Coatings are listed in order of preparation in the case of multiple coatings.

(b) Total Xe^{133} in ppm released at maximum temperature of 1300 C after neutron activation, except for Lot 717B, which was heated to only 1100 C.

Work This Period

For the purpose of incorporating oxide-coated particles in oxide matrices, it is desirable to have metal oxide coatings which can be heated above 1400 C without grain growth and associated loss of coating strength and integrity. Consequently, work has been started on the codeposition of a grain-growth inhibitors.

Carbon-Coating Studies

A. C. Secrest, V. M. Secrest,
M. F. Browning, and J. M. Blocher, Jr.

Objective

To establish conditions for coating fuel particles with pyrolytic carbon of optimum quality.

Program Approach

Pyrolytic-carbon coatings are being applied by the decomposition of hydrocarbons in a fluidized bed of particles at temperatures of 950 to 2000 C. The coatings are being evaluated by means of microscopic examination, measurement of physical properties, direct alpha counting, and by measurement of fission-gas release on postirradiation heating. Promising materials will be evaluated by in-pile tests at elevated temperatures.

Progress Reported Previously

Pyrolytic carbon coatings have been applied to fuel particles at temperatures of 950 to 1800 C. In addition, normal uranium shot (74 to 149 μ) was coated with 27 μ of PyC at 600 to 1400 C, heated 5 hrs at 1500 C in helium, and overcoated with 31 μ of PyC at 1400 C. A PyC-coated UC₂ product resulted which exhibited a postirradiation Xe¹³³ release at 1300 C of only 1.3 ppm.

The carbon-coated enriched fuel particles listed in the following tabulation have been submitted for in-pile evaluation:

Lot	Fuel Particle		Coating Temperature, C	Coating Description ^(a)	Xenon Release Ratio ^(b) , ppm
	Type	Diameter, μ			
918A	UC ₂ -E1	200	1400	55 μ of columnar PyC	5.0
919A	UC ₂ -E1	200	1400	55 μ of laminar PyC	<0.5
920A	UO ₂ -E3	127	1400	45 μ of laminar PyC	0.75
921	UO ₂ -E2	127	1000	48 μ of dense Al ₂ O ₃	0.5
		1	1400	32 μ of laminar-columnar PyC	
922	UO ₂ -E3	127	1000	8 μ of dense Al ₂ O ₃	3.1
			700	18 μ of porous Al ₂ O ₃	
			1000	28 μ of dense Al ₂ O ₃	
			1400	65 μ of laminar-columnar PyC	
966C	UC ₂ -E2	127	950	44 μ of laminar PyC	0.08
1010A	UO ₂ -E3	127	1300	11 μ of laminar PyC	1.3
			1600	55 μ of laminar PyC	

(a) Coatings are listed in order of preparation in the case of multiple coatings.

(b) Total Xe¹³³ in ppm release at maximum temperature of 1300 C after neutron activation.

Work This Period

Carbon-coated fuel particles have consistently failed after a few weeks in-pile. The cause of failure is not known, but radiation distortion of the fuel and/or coating may well be a factor. Consequently, a program has been initiated to develop a PyC-coated particle with a void beneath the outer coating which would accommodate this distortion.

A new lot of coated fuel particles (Lot 973) is being prepared with an enriched UO_2 substrate and provision for void formation. The coating is to consist of a 25- μ inner layer of sinterable porous Al_2O_3 , 0.5 to 1 μ of dense Al_2O_3 to seal the porous Al_2O_3 , and 30 to 40 μ of PyC to form the pressure vessel. When heated, the porous Al_2O_3 is expected to shrink, leaving a void adjacent to the PyC. The preparation of this material is nearly completed. Its void-forming characteristics will be investigated immediately along with its performance in the standard evaluation tests.

Plans for Future Work

The program to design and prepare distortion-accommodating PyC-coated particles will be continued.

Beryllia-Coating Studies

W. J. Wilson, V. M. Secrest,
M. F. Browning, and J. M. Blocher, Jr.

Objective

To develop procedures for coating fuel particles with BeO of optimum quality.

Program Approach

Beryllia is being deposited on UO_2 shot by hydrolysis of BeCl_2 in a fluidized bed. The use of other compounds from which BeO can be formed will also be explored. The coatings are being evaluated by microscopic examination, physical-property determinations, alpha counting, and measurement of fission-gas release on postirradiation heating. Promising materials will be evaluated by in-pile tests at elevated temperatures.

Progress Reported Previously

Beryllia coatings have been deposited primarily by the steam hydrolysis of BeCl_2 at temperatures of 800, 1000, 1100, 1300, and 1400 C. In general, all BeO coatings have contained varying degrees of porosity. In addition the coatings which contained significant quantities of impurities were the most dense and nonporous. All coatings prepared by the oxidation or decomposition of beryllium basic acetate were contaminated with carbon, contained hydroxy residue, and were soluble in nitric acid. A sample designated Lot 835C was prepared at 1400 C on depleted urania by the steam hydrolysis of BeCl_2 and exhibited an encouraging postirradiation Xe^{133} release of 17 ppm. Last month Lot 843C was prepared with depleted UO_2 at approximately 1300 C, and had a BeO coating thickness of 54 μ .

Work This Period

Coating of enriched UO_2 shot with BeO by steam hydrolysis of BeCl_2 at 1400 C yielded Lot 845E. The coating is $60\ \mu$ thick, is dense and transparent, and shows a hardness in the range of 1080 to 1300 KHN, which is essentially equal to that of single-crystal BeO . Particles $280\ \mu$ in over-all diameter show an average crushing strength of 875 g. In the standard leach test Lot 845E lost 10 ppm of UO_2 . After heating in air at 1100 C for 20 hr, the standard leach test resulted in a loss of less than 10 ppm of UO_2 . Heat treatment of a neutron-activated sample of Lot 845E yielded a total of 0.10-ppm xenon-133 release at temperatures up to 1300 C. These results were sufficiently encouraging to warrant in-pile radiation studies which are now under way.

Evaluation of Lot 843C was continued. The loss of 80 ppm of UO_2 in the standard leach test of as-coated particles was encouraging. However, after being heated in air at 1100 C for 20 hr the material lost 1780 ppm UO_2 in the standard leach test. Further evaluation was suspended.

Following the preparation of Lot 845E the coating equipment was altered to improve its operation. In the new system BeCl_2 is formed as required in the lower portion of the Mullite reactor tube by reacting beryllium with HCl . The BeCl_2 formed is carried by the main hydrogen stream through a preheater section into the fluidized bed as before. The hydrolysis agent is obtained by the reaction of CO_2 and hydrogen which are passed into the bed through the center tube.

This system eliminates bothersome heated ball joints and vapor-transfer lines, and should permit more rapid turnaround of equipment and better control of reactant composition. The system performed well in a preliminary run.

Plans for Future Work

Further coatings will be made in the new system to improve both the reliability of operation and also the uniformity of the product, both from particle to particle in the product of a single run, and from one run to the next.

Additional Oxide-Coating Studies

M. F. Browning, V. M. Secrest, and J. M. Blocher, Jr.

Objective

To explore briefly the potential of MgO , Ce_2O_3 , and other oxides as coatings for fuel particles.

Program Approach

Magnesia is being deposited on UO_2 particles by hydrolysis of MgI_2 vapor in a fluidized bed. The coatings are being evaluated by microscopic examination, measurement of physical properties, alpha counting, and measurement of fission-gas release on postirradiation heating. Promising materials will be evaluated by in-pile irradiation at elevated temperature.

Progress Reported Previously

Magnesia coatings were deposited at 800, 1100, and 1400 C by hydrolysis of MgI_2 fed as granules directly into the fluidized bed of particles to be coated. Coatings of magnesia deposited at 1400 C were relatively transparent and appeared to be dense and free of voids. Hardness was essentially equal to that of fused MgO . The lower temperature coatings were less transparent and appeared to contain submicron pores. In general, MgO coatings on a UO_2 substrate were found by microscopic examination to be cracked when cooled to room temperature. It was concluded that this cracking resulted from the large difference (4×10^{-6} per C) in the thermal-expansion coefficients of UO_2

and MgO. Cracking of the MgO coating was reduced but not eliminated by using a porous UO_2 substrate. A void was provided between the substrate and coating which reduced cracking of the MgO coating. However, sufficient voidage was not maintained in every particle during processing, so cracking was not completely eliminated.

Work This Period

A sample of dense UO_2 coated with $40\ \mu$ of dense MgO at 1400 C (Lot 1116) was heated in oxygen, and its weight change was recorded on a microbalance. The sample was heated for 12 hr at 800 C. The only significant change was an increase in weight at about 700 C equivalent to approximately 1 per cent of the particles. The particles which oxidized at this temperature are thought to be those which were cracked in the as-produced form. At 1100 C the sample increased in weight uniformly until, after 40 hr, approximately 60 per cent of the UO_2 in the sample was oxidized. Then the temperature was increased to 1300 C where only 2.5 hr was required to oxidize the remaining UO_2 . The low, steady rate of oxidation at 1100 C is taken to indicate that oxidation occurs by diffusion through the coating rather than through cracks formed by heating the particles above a certain minimum temperature. This will be confirmed by briefly heating the particles to a temperature above that at which oxidation is detectable and then returning to the original temperature to determine if the initial rate of oxidation is re-established.

Plans for Future Work

Except for an experiment to confirm the mode of oxygen transfer, experimental work on MgO coating will be deferred until promising methods of compensating for the thermal-expansion difference and of preparing coatings more resistant to oxygen diffusion are devised.

Chemical Fabrication Studies

A. C. Secrest, M. F. Browning, V. M. Secrest,
and J. M. Blocher, Jr.

Work on this phase has been indefinitely suspended.

Graphite-Matrix Studies

M. C. Brockway, D. J. Bowers, and W. H. Duckworth

Objective

To establish fuel dispersion methods and fueled-graphite processing conditions that will provide acceptable graphite-matrix quality without damaging dispersed coated-particle fuels, and to prepare and characterize specimens of such fuel elements for irradiation testing.

Program Approach

Major effort is being devoted to preparing satisfactory random dispersions of each type of coated-particle fuel in matrices possessing properties comparable to present reactor-grade graphites. Also, the preparation of equidistant fuel dispersions and the use of matrices having special properties are being explored to determine irradiation effects of these variables. Specimen characterization includes obtaining pertinent physical properties, radiographic and metallographic examination of fuel dispersion, non-destructive determination of fuel loading by gamma emission, and evaluation of coating integrity by neutron-activation tests.

Progress Reported Previously

A batch method was developed for randomly dispersing Al_2O_3 -coated UO_2 particles in graphite by conventional processing except for the use of solvent mixing. A summary of characterization data is given in BMI-1549 for three sets of specimens prepared by this method and selected for static capsule irradiation.

To meet more effectively the objectives of this program, specimen-quantity random-dispersal methods were developed and used in combination with pressure-baking procedures similar to those outlined in GAMD-2221. Thirteen types of coated enriched fuel particles (Lots 4E, 918A, 919A, 920A, 725A, 1010A, 715A-HT, 732D, 733B, 734C, 736B, 935C, and 966C) were incorporated into fuel elements by these methods. Following neutron activation, specimens containing these particles released about the same or lesser amounts of fission gas at 1150 C than did the respective undispersed coated particles at 1300 C.

A method of pelletizing individual coated fuel particles with matrix material has led to improved dispersion of fuel in graphite. Through the use of this technique, dispersions in fuel elements have been prepared which approximate equidistant fuel-particle separation. Also, fuel-element specimens containing Al_2O_3 -coated UO_2 particles have been molded at pressures up to 40,000 psi without fuel-particle damage as checked by neutron-activation tests. High molding pressures yield high matrix strength and density.

Work This Period

A set of fueled-graphite specimens was prepared to evaluate molding-pressure damage which can occur in randomly dispersed PyC-coated UC_2 . Lot 955C fuel was randomly dispersed (solvent mixing in specimen quantity) in a graphite-binder mix similar to one which has been used in pelletizing Al_2O_3 -coated fuel. Molding pressures of 5,000, 10,000, 20,000, and 40,000 psi were used at 80 ± 5 C. The resulting fueled cylinders were pressure baked at 5,000 psi to a temperature of 1250 C. They then were neutron activated. Xenon-release data, given in Table L-3, indicated that damage of the randomly dispersed fuel resulted at all of the molding pressures used. As expected, the higher molding pressures produced greatly increased fuel damage.

TABLE L-3. XENON-133 RELEASE^(a) AFTER NEUTRON ACTIVATION OF GRAPHITE FUELED WITH RANDOMLY DISPERSED LOT 966C FUEL

Molding Pressure, psi	Xenon-133 Release, ppm
5,000	24.5
10,000	13.2
20,000	455
40,000	1390

(a) Release values obtained in heating 1 hr at 1150 C. For comparison, undispersed Lot 966C fuel gave 0.08 ppm xenon-133 release at 1320 C.

When the Lot 966C fuel has been pelletized and made into fueled specimens, the above release data will permit a quantitative assessment of the protection against molding damage which pelletizing can provide this specific fuel.

Because a relatively isotropic carbon matrix exhibits radiation-induced shrinkage, it might provide better mechanical support to coated-fuel particle than does a graphite matrix. Accordingly, attention is being given to the selection of a carbon-binder combination which can be used to pelletize fuel and which can then be suitably formed and baked. Unfueled specimens have been prepared from carbons of different types, particle sizing, and binder (pitch) contents. When mechanical properties have been obtained for specimens prepared from these compositions, one or more will be selected for pelletizing study.

Plans for Future Work

New fuel types will be dispersed in graphite by pelletizing techniques. Attention will be given also to carbon matrices, or to graphite matrices having special properties.

Pressure-Bonding Studies

S. W. Porembka, E. S. Hodge, and S. J. Paprocki

This program has been temporarily suspended.

FUEL-IRRADIATION PROGRAM

W. H. Goldthwaite, D. N. Sunderman, and R. F. Dickerson

Sweep Capsule CFP-S-3

N. E. Miller, W. J. Zielenbach, R. L. Ritzman, and R. J. Burian

Objective

To rapidly and economically obtain fission-gas-release information on a new type material, beryllia-coated uranium dioxide particles, while it is being irradiated and maintained at approximately 100 C.

Program Approach

Unsupported particles of Lot 845E, encapsulated in a small, noninstrumented, sweep capsule, will be irradiated in the core of the BRR and will operate at approximately 100 C. A stream of helium will continually flow past the specimen to sweep the release fission gases to an out-of-pile analytical system. The experiment will be conducted for 2 weeks.

Work Reported Previously

This experiment was planned in the initial stages of this program to be the first experiment with beryllia-coated fuel and was to operate at elevated temperatures to a relatively high burnup. With this type material just being made available late in June, the objective of the experiment was changed to obtain more rapid information.

Work This Period

The assembly of the capsule was completed and the experiment was begun June 27; no data are available as yet.

Plans for Future Work

This irradiation will be terminated July 9. There is no postirradiation examination planned. The data from the fission-gas-samples will be reported next month.

Sweep Capsule CFP-S-10

N. E. Miller, W. J. Zielenbach, R. L. Ritzman, and R. J. Burian

Objective

To obtain continuous fission-gas-release information from dense uranium dioxide particles, coated with an inner layer of pyrolytic carbon and an outer layer of alumina, during irradiation at 1050 C to a burnup approaching 50,000 MWD/Tonne and to obtain postirradiation information on several other new coated fuel materials.

Program Approach

The sweep fuel specimen is contained in a separate compartment of the capsule. During the irradiation a helium sweep gas continuously flows past the specimen, carrying the released fission gases to an out-of-pile sampling system where they may be collected for analysis. In addition, five individually canned static specimens are contained in the static section of the capsule. During the irradiation the fuel specimens will be maintained at a nominal temperature of 1050 C.

Progress Reported Previously

The sweep specimen consists of two 1/4-in.-diameter by 1/4-in.-long graphite-matrix pellets containing dense UO_2 particles coated with a layer of pyrolytic carbon and a layer of alumina, Lot 736B. The static specimens are unsupported coated particles from Lots 725D, 735C, and 966C and graphite-matrix elements containing coated particles from Lots 732D and 736B. This experiment was begun on May 2, 1962, with the sweep specimen operating at approximately 1050 C. During the first week of operation, three of the four auxiliary electrical capsule heaters failed, and the operating temperature was reduced to approximately 800 C. Through the first reactor cycle, the fission-gas-release fractions remained relatively constant in the order of 10^{-8} . During the third week of operating, the release rate began to increase so that at the end of the second cycle in late May the release fractions were in the order of 10^{-5} .

Work This Period

This experiment has been continued through June with operating temperatures between 700 and 800 C. The fission-gas-release data calculated from adsorption samples are given in Table L-4.

TABLE L-4. FISSION-GAS-RELEASE DATA FROM SWEEP CAPSULE CFP-S-10

Sample	Accumulated Irradiation Time, hr	R/B				
		Kr ⁸⁷	Kr ⁸⁸	Xe ¹³⁵	Kr ^{85m}	Xe ¹³³
10-1A	1	--	--	3.3×10^{-9}	7.6×10^{-8}	--
10-2A	42	3.2×10^{-8}	2.1×10^{-8}	2.5×10^{-8}	5.5×10^{-8}	5.6×10^{-8}
10-3A	130	--	1.5×10^{-8}	1.1×10^{-8}	3.1×10^{-8}	3.1×10^{-8}
10-4A	175	--	--	2.7×10^{-8}	2.5×10^{-8}	5.6×10^{-8}
10-5A(a)	269	--	--	--	--	--
10-6A	286	--	--	2.6×10^{-8}	1.8×10^{-8}	3.2×10^{-8}
10-7A	359	4.2×10^{-6}	2.6×10^{-6}	5.4×10^{-6}	8.0×10^{-6}	4.5×10^{-5}
10-8A(b)	370	1.2×10^{-5}	5.0×10^{-6}	9.3×10^{-6}	1.5×10^{-5}	4.2×10^{-5}
10-9A	421	6.9×10^{-6}	3.7×10^{-6}	2.3×10^{-5}	1.4×10^{-5}	4.4×10^{-4}
10-10A	445	1.4×10^{-5}	6.7×10^{-6}	1.7×10^{-5}	2.3×10^{-5}	1.6×10^{-4}
10-11A	516	4.8×10^{-5}	2.9×10^{-5}	5.1×10^{-5}	5.4×10^{-5}	1.3×10^{-4}
10-12A(c)	516	--	--	4.2×10^{-6}	--	5.3×10^{-6}
10-13A	809	--	3.0×10^{-4}	1.2×10^{-3}	8.7×10^{-4}	3.6×10^{-3}
10-14A(b)	820	1.1×10^{-3}	3.3×10^{-4}	5.2×10^{-4}	6.3×10^{-4}	1.5×10^{-3}

(a) Taken 1.5 hr after shutdown. No fission-gas activity.

(b) R/B value for Kr⁸⁷ uncertain by a factor of about two.

(c) Taken 1.5 hr after shutdown. Presence of krypton activity possible.

The early fission-gas release is attributed to recoils escaping from slight uranium contamination in the particle coatings, since it is nearly half-life independent. The increase in release beginning with Sample 10-7A indicates progressive coating failure such that at Sample 10-14A most of the coatings are probably cracked. The relative R/B values for Kr⁸⁷ and Kr⁸⁸, in which Kr⁸⁷ is higher than Kr⁸⁸ despite the shorter half-life for Kr⁸⁷, indicate that the bromine precursors contributed to the measured rare-gas release. The apparent minor amounts of krypton activity in Sample 10-12A, which was taken 1.5 hr after shutdown, also supports this observation.

Plans for Future Work

This irradiation is scheduled to be terminated July 23, 1962.

Postirradiation Studies

R. J. Burian, N. E. Miller, R. L. Ritzman, and J. E. Gates

Objective

To evaluate the effects of reactor radiation on various types of coated particles as a function of fuel and coating materials, porosity, coating fabrication techniques, irradiation temperature, flux, and matrix support.

Program Approach

Various types of coated particles are being evaluated after irradiation to determine the fission-product-retention properties of the coatings. Different combinations of pyrolytic carbon and high- and low-density Al_2O_3 coatings have been deposited on high- and low-density UO_2 and UC_2 particles. Parameters being studied include the size, porosity, and material of the fuel particle, the thickness, porosity, and material of the coating, and the effect of heat treatment before irradiation, irradiation temperature, fuel burnup, and support from a matrix on the performance of the coatings.

The coated particles are being irradiated in either a static helium atmosphere or in a dynamic helium atmosphere whereby gases released from the particles are purged from the capsule, collected, and analyzed to obtain in-pile fission-gas-release data. Postirradiation studies include analyses of samples of gases released from the particles irradiated in a static helium atmosphere, leaching of irradiated specimens with nitric acid to determine the number of particles with failed coatings, heat treatment to determine fraction of gas retained in particles with failed coatings, and metallographic examination to study structural changes in the particles.

Progress Reported Previously

Fifty-four specimens comprising 12 types of coated particles have been irradiated. All but 5 of these specimens were removed from the capsules after irradiation and examined. The results of these examinations and an evaluation of particle performance have been presented in previous reports by capsule number. Henceforth, to aid the reader in evaluating the performance of the coated-particle specimens, postirradiation data of all specimens will be presented in this section. The results of previous postirradiation examinations of all specimens are presented in Table L-5.

The 54 specimens were irradiated at temperatures of about 100 or 1070 C to burn-ups ranging up to 5.0 per cent of the uranium. The results of the postirradiation gas-release studies indicate, in general, that dense Al_2O_3 coatings experience nearly complete failure at 100 C but perform much better at 1070 C. Coatings with a porous Al_2O_3 layer between two dense Al_2O_3 layers perform slightly better at 100 C than fully dense Al_2O_3 coatings, but their performance does not improve at the elevated temperature. Studies indicate that there is not a significant improvement in gas-retention ability of the particles dispersed in a graphite matrix. The results of analyses of acid solutions used to leach the specimens indicate in a number of cases a greater degree of coating failure than was indicated by the postirradiation gas-release studies. An investigation into the source of the lack of agreement between these two techniques was initiated.

The results of postirradiation examinations of gas release from pyrolytic-carbon-coated particles are less consistent than data from examinations of alumina-coated particles. However, in general, the data indicate that pyrolytic-carbon coatings fail more severely than Al_2O_3 coatings at both 100 C and 1070 C.

Work This Period

Six specimens irradiated in Capsule CFP-S-8 were removed from the specimen cans. However, in the case of two specimens of unsupported particles, very few

TABLE L-5. SUMMARY OF RESULTS OF POSTIRRADIATION EXAMINATION OF COATED-PARTICLE SPECIMENS

Lot	Fuel Material	Coating ^(a)		CFP Irradiation Capsule	Matrix Support ^(b) ,	Irradiation Temperature, C	U ²³⁵	Fractional	Uranium
		Material	Thickness, μ				Fission Burnup, per cent	Xe ¹³³ Release, ppm	Removed in Leach, per cent
<u>Specimens With Alumina Coatings</u>									
4E	UO ₂	d-Al ₂ O ₃	8						
		p-Al ₂ O ₃	6						
		d-Al ₂ O ₃	19	4	None	100	2.9	10	28
				S-5	Graphite	1070	3.9	0.3	--
715A	UO ₂	d-Al ₂ O ₃	42	1D	None	100	3.3	120	54
				1B	None	100	4.7	140	39
				4	None	100	3.0	120	--
				S-7	None	100	3.9	60	--
				1C	None	1070	1.7	0.1	0.9
				1A	None	1070	3.2	0.02	0.2
				1D	Graphite	100	3.5	2	33
				1B	Graphite	100	5.0	0.8 ^(c)	43.38 ^(c)
				1C	Graphite	1070	1.7	--	--
				1A	Graphite	1070	3.3	4	--
715A-HT	UO ₂	d-Al ₂ O ₃	42	S-7	None	100	3.9	0.01	95
				S-8	None	1070	--	--	--
				S-7	Graphite	100	3.6	0.5	93
				S-8	Graphite	1070	--	--	--
717B	UO ₂	d-Al ₂ O ₃	43	S-7	None	100	3.9	--	--
				S-8	None	1070	--	--	--
721C	UO ₂	d-Al ₂ O ₃	8						
		p-Al ₂ O ₃	18						
		d-Al ₂ O ₃	28	1D	None	100	3.4	2	88
				1B	None	100	4.6	20	5
				4	None	100	3.1	30	95
				1C	None	1070	1.7	20	3
				1A	None	1070	3.2	4	0.5
				1D	Graphite	100	3.4	20	44
				1B	Graphite	100	4.8	2	30
				1C	Graphite	1070	1.7	40	--
				1A	Graphite	1070	3.4	150	--
				1D	Carbon	100	3.0	2	33
				1B	Carbon	100	4.9	4	43
				1C	Carbon	1070	1.6	2	--
				1A	Carbon	1070	3.3	60	--
725D	UO ₂	d-Al ₂ O ₃	56	S-7	None	100	3.9	20	--
				S-6	None	1070	3 (est.)	700	--
				S-6	Graphite	1070	3 (est.)	30 ^(d)	--
733B	UO ₂	d-Al ₂ O ₃	47	S-7	None	100	3.9	--	--
				S-8	None	1070	--	--	--

TABLE L-5. (Continued)

Lot	Fuel Material	Coating ^(a)		CFP Irradiation Capsule	Matrix Support ^(b)	Irradiation Temperature, C	U ²³⁵ Fission	Fractional Xe ¹³³	Uranium Removed
		Material	Thickness, μ				Burnup, per cent	Release, ppm	in Leach, per cent
Specimens With Pyrolytic-Carbon Coatings									
918A	UC ₂	Col PyC	55	4	None	100	3.1	250	93
				S-5	None	1070	3.8	6200	21
919A	UC ₂	Lam PyC	55	4	None	100	3.2	20	--
				S-5	None	1070	3.7	9000	55
				S-6	Graphite	1070	3 (est.)	30 ^(e)	--
920A	UO ₂	Lam PyC	45	4	None	100	3.4	290	30
				S-7	None	100	3.7	80	--
				S-5	None	1070	3.8	2100	66
				S-6	Graphite	1070	3 (est.)	400	--
921	UO ₂	d-Al ₂ O ₃	48	1D 1B 1C 1A	None	100	3.1	60	46
		Mix PyC	32			100	4.9	20	2.2
						1070	1.6	0.2	7.2
						1070	3.3	0.02	3.0
1010A	UO ₂	Lam PyC	66	S-7	None	100	3.6	--	--
				S-8	None	1070	--	--	--

(a) In order of deposition. Code used is as follows:

d-Al₂O₃: Essentially fully dense Al₂O₃ layer

p-Al₂O₃: Relatively porous Al₂O₃ layer

Col PyC: Pyrolytic carbon with a columnar or radially oriented structure

Lam PyC: Pyrolytic carbon with a laminar or circumferentially oriented structure

Mix PyC: Pyrolytic carbon with a mixed columnar and laminar structure.

(b) Three conditions of matrix support existed:

(1) None: no support from a matrix.

(2) Graphite: support afforded by a relatively graphitized matrix.

(3) Carbon: support afforded by a relatively ungraphitized matrix.

(c) This specimen consisted of two fueled-graphite pellets. Leach of one pellet as recovered from the capsules yielded 43 per cent uranium recovery. Second pellet was heated to 815 C at which temperature 270 ppm of Kr⁸⁵ was driven off. Subsequent leach of second pellet yielded 38 per cent uranium recovery.

(d) This specimen consisted of two fueled-graphite pellets. Analysis of gases sampled from within the specimen can after irradiation indicated 30 ppm of Xe¹³³ was released by both pellets. Subsequent heating of one pellet to 540 C drove off an additional 1.4 ppm of Xe¹³³.

(e) This specimen also consisted of two pellets. Room-temperature release of both pellets was 30 ppm Xe¹³³. Subsequent heating of one pellet to 540 C drove off an additional 30 ppm Xe¹³³.

particles could be recovered. Also, one matrix specimen could not be removed from the specimen can by the procedures previously used. The analyses of gas samples from the five static specimen cans irradiated in this capsule were completed. Evaluation of the results is in progress.

Metallographic examination of four specimens is in progress. The results obtained to date reveal that a second phase is present in the UO_2 of all alumina-coated particles which were observed to have failed coatings. This second phase was not observed in the UO_2 of any particles with intact Al_2O_3 coatings. Evidence of plastic flow of the UO_2 was observed in particles irradiated both at 100 and 1070 C. This phenomenon has previously been reported in the case of UO_2 cermet and UO_2 - ZrO_2 fuels. The metallographic examinations are continuing.

Studies of the source of discrepancy between leach data and postirradiation gas-release data are in progress.

Plans for Future Work

Evaluation of gases sampled from the five static specimen cans from Capsule CFP-S-8 will be completed. Specimens recovered from this capsule will be leached and/or heat treated. Efforts will be directed toward identifying the second phase observed in the UO_2 of particles with failed Al_2O_3 coatings. Additional specimens will be examined metallographically.

Intermediate-Temperature Design Studies

N. E. Miller and W. J. Zielenbach

This work is currently recessed.

Fission-Gas Analytical Studies

R. L. Ritzman and T. S. Elleman

This program was recessed during June.

SUPPORTING RESEARCH PROGRAM

Radiation-Damage Studies

T. G. Knorr and C. M. Schwartz

Objective

To study individual fission-recoil particle effects in nonfissionable ceramics by characterizing the damage they produce in the surface and volume of the material.

Program Approach

Replica and decoration procedures are being investigated to allow observation of the fission tracks by means of electron or optical microscopy. Alpha Al_2O_3 and other ceramics will ultimately be studied, but the alkali halides will be investigated initially because of ease of handling and the information already available. Fresh cleavage faces are fueled by contact with a uranium-bearing material during irradiation. Replicas can then be made by shadowing the damaged faces with platinum at an angle of about 20 deg and depositing carbon normally. These replicas can be floated off in an appropriate liquid and observed with transmission electron microscopy. In the case of decoration techniques, optical microscopy will be used where possible.

Progress Reported Previously

Direct platinum-carbon replica techniques have revealed fission-recoil damage on the surfaces of NaCl, KBr, and LiF, as described in summary in a recent progress report (BMI-1565). Decoration by the precipitation of silver in AgCl has also displayed fission tracks in this material. Humidity and/or temperature have been found to affect the register of the events in NaCl to the point where they have become actually unobservable in some specimens. A detailed analytical study of these data was initiated in an attempt to determine the most likely cause of the observed damage and the effects produced by the various atmospheric conditions.

Work This Period

The continuing analysis of the size distribution in LiF indicates that the hardening mechanism due to gamma irradiation may not be as significant as was stated last month. Currently, it appears that radiation-enhanced diffusion might be relatively more important than originally thought. In an attempt to try to distinguish between some of the various possibilities, a series of samples was irradiated under different conditions of gamma and neutron dose but designed to have the same fission flux. No results are available as yet.

Plans for Future Work

The analytical study will be continued.

Fission-Gas-Diffusion Studies

D. L. Morrison, R. H. Barnes, T. S. Elleman,
and D. N. Sunderman

Objective

To determine diffusion coefficients in the absence of radiation for rare-gas fission products in alumina, pyrolytic carbon, and other potential coating materials for nuclear fuel particles.

Program Approach

Small particles of the coating material of interest are packed in uranium dioxide powder and irradiated at low temperatures in the Battelle Research Reactor. These particles, which contain fission products recoiled into them from the surrounding uranium dioxide powder during irradiation, are separated from the powder and heated at constant temperature in a flowing helium stream. The fission gases released from the particles during heating are collected from the helium on activated charcoal at liquid-nitrogen temperature and radioassayed. The results of the measurements are then used to determine fission-gas-diffusion coefficients.

Progress Previously Reported

Diffusion coefficients for xenon-133 in Linde Company 1000- μ -diameter alpha-alumina spheres have been measured in the temperature range 500 to 1550 C as a function of fission-fragment recoil exposure. Similar measurements have been made with laminar-type pyrolytic-carbon coatings. The results of these postirradiation release measurements were summarized in BMI-1569 and BMI-1581. Xenon-133 release measurements on columnar-type pyrolytic carbon coatings were reported in BMI-1574, and similar measurements on single-crystal beryllia particles were reported last month in BMI-1581.

Work This Period

Single-crystal magnesia specimens, both pure and doped, which were obtained from the Norton Company in the form of rectangular parallelepipeds, were irradiated in contact with natural uranium-metal foil for 1 week in a flux of 6×10^{12} nv. Three types of doped crystals were used: magnesia doped with iron, nickel, and zirconium to impurity concentrations of 0.05, 0.03, and 0.03 mole per cent, respectively. Postirradiation heating experiments were performed on these specimens. The results of these experiments are summarized in Table L-6. Data on release measurements from alpha-alumina spheres with an exposure of 3×10^{13} fission-product recoils per cm^2 are included for comparison.

TABLE L-6. SUMMARY OF XENON-133 RELEASE DATA FOR MAGNESIA CRYSTALS

Sample	Temperature, C	Fraction of Xenon-133 Released During Initial Burst	Total Fraction of Xenon-133 Released During 315-Min Heating	Diffusion Coefficient, cm^2 per sec
Al_2O_3	800	7.5×10^{-4}	1.0×10^{-3}	1.0×10^{-18}
	1000	2.0×10^{-3}	7.0×10^{-3}	2.6×10^{-17}
	1200	1.0×10^{-2}	1.5×10^{-2}	8.0×10^{-16}
	1500	8.0×10^{-2}	1.1×10^{-1}	4.0×10^{-14}
MgO, pure	800	6.4×10^{-4}	8.5×10^{-4}	5.2×10^{-19}
	1000	1.2×10^{-3}	1.8×10^{-3}	3.3×10^{-18}
	1200	2.2×10^{-2}	3.2×10^{-2}	1.5×10^{-15}
	1500	--	1×10^0	--
MgO, Fe_2O_3 -doped	800	2.3×10^{-3}	4.5×10^{-3}	4.5×10^{-19}
	1200	1.7×10^{-2}	2.6×10^{-2}	1.4×10^{-15}
	1500	--	1×10^0	--
MgO, NiO-doped	800	4.0×10^{-4}	5.6×10^{-4}	2.8×10^{-19}
	1200	7.0×10^{-3}	8.2×10^{-3}	1.5×10^{-17}
MgO, ZrO_2 -doped	1200	1.5×10^{-2}	2.2×10^{-2}	4.3×10^{-16}

Diffusion coefficients were calculated for the magnesia and are compared in Table L-6 with the average values for the diffusion coefficients in alumina. In general, the postirradiation release of xenon-133 from the four types of magnesia specimens studied is comparable to that from alpha alumina up to temperatures of 1200 C. However, the release from magnesia at 1500 C is much greater than from alumina, with essentially all of the xenon-133 being released during the 315-min heat treatment. The accumulative fractional release at 1500 C was not linear with the square root of the heating time so no diffusion coefficient was calculated at this temperature.

Plans for Future Work

Diffusion experiments are to be performed on other potential coating materials including TiO_2 and stabilized ZrO_2 . In addition to the diffusion measurements on potential coating materials, postirradiation measurements are planned for actual alumina coatings.

Fuel-Compatibility Studies

G. W. Cunningham, D. C. Carmichael, R. H. Barnes,
and S. J. Paprocki

Objective

To develop techniques and investigate reactions which may affect the maximum operating time or temperature of coated fuel particles at temperatures to 1800 C.

Program Approach

The stability of oxide-coated UO_2 particles in an oxidizing atmosphere will be studied by measuring weight changes and by determination of changes in oxygen content of the UO_2 in the temperature range of 1000 to 1800 C. Specimens will be examined by metallographic and X-ray diffraction techniques, and the results will be compared with weight-change data obtained on a recording microbalance. Studies on the oxide-carbon reactions will involve the use of gravimetric and metallographic techniques to determine reaction kinetics of the oxides with pyrolytic carbon. Diffusion of uranium through pyrolytic-carbon coatings on UC, UC_2 , and UO_2 particles will be investigated by annealing coated particles of fully enriched uranium compounds at temperatures of 1000 to 1800 C for various times. The distribution of uranium in the carbon coating will then be determined by a combination of metallographic, autoradiographic, and alpha-counting techniques. The redistribution of uranium due to a thermal gradient will be studied by annealing specimens of carbon-coated UC_2 in a temperature gradient for specified periods of time.

Progress Reported Previously

Results have been presented in previous progress reports for various compatibility studies with Al_2O_3 -coated UO_2 (BMI-1557), carbon-coated UO_2 (BMI-1514 and BMI-1518), carbon-overcoated Al_2O_3 -coated UO_2 (BMI-1534), and carbon-coated UC (BMI-1534). A topical report is being prepared on the oxidation of Al_2O_3 -coated UO_2 particles.

Autoradiographic and alpha-counting techniques have been studied for measuring the rate of uranium diffusion through pyrolytic-carbon coatings on enriched, spherical UC_2 particles. Equations for interpreting the results of the measurements in terms of diffusion coefficients, assuming uniform diffusion, were presented in BMI-1561. Samples of the particles with two types of pyrolytic-carbon coatings, laminar and columnar, were vacuum heat treated at 1600 C for 0.5 and 2 hr. The alpha-counting measurements of the samples indicated that uranium diffuses more rapidly through the columnar than

through the laminar coatings. The diffusion coefficient calculated using autoradiographic data for the particles with the laminar coatings heated at 1600 C was $5.6 \times 10^{-10} \text{ cm}^2 \text{ per sec.}$

It was found that the production of Al_4C_3 as a result of the reaction between Al_2O_3 and carbon at 1400 and 1500 C is not the rate-controlling process and that there is a volatile product, probably a suboxide, formed in the reaction. Quantitative results at 1400 and 1450 C were reported last month in BMI-1581.

Work This Period

The study of the rate of uranium diffusion through pyrolytic-carbon coatings on UC_2 particles by use of autoradiographic and alpha-counting techniques has continued. Using an experimental NTB film and improved techniques recently developed, autoradiographs were made of the particles with laminar- and columnar-type coatings which had been heat treated in vacuum at 1600 C for periods of 0.5, 1, and 2 hr. Analysis of the data from these autoradiographs is currently in progress.

Work on the thermal diffusion of uranium in carbon-coated UC_2 particles is continuing. Autoradiographic examination of particles of this type heated in a thermal gradient at 1500 C have revealed no evidence of thermal migration.

Samples of carbon-coated materials containing UO_2 and oxygen contaminant are being prepared for thermal-diffusion studies.

Plans for Future Work

The analysis will be completed of the autoradiographic data obtained for the carbon-coated UC_2 particles which were heated at 1600 C for the various times.

In the study of thermal diffusion of uranium in carbon-coated fuel particles, the coated UO_2 samples will be heated in a thermal gradient and evaluated to study the possible effect of the presence of some oxide in coated UC_2 particles.

Tests will be run to determine the effect of moisture on the oxidation resistance of Al_2O_3 -coated UO_2 particles.

Long-time tests (1,000 to 10,000 hr) are being planned to determine the stability of oxide-coated particles in an oxygen atmosphere and the reaction rate of oxide particles with a carbon matrix in the presence of various partial pressures of CO.

Coolant-Compatibility Studies

C. A. Alexander, J. S. Ogden, A. Levy, and R. C. Himes

Objective

To establish experimentally the compatibility of high-pressure steam with Al_2O_3 , MgO , ZrO_2 , and UO_2 , with respect to erosive losses which impair their use in a nuclear reactor.

Program Approach

A transpiration study is being conducted in which the effect of steam on the rate of volatilization is under investigation. Initial investigations are being carried out at low steam partial pressures; under these conditions vaporization mechanisms may be more precisely determined.

Progress Reported Previously

Results of a literature study have been issued as BMI-1530. Experimental data on the effect of water vapor on UO_2 volatilization, and thermodynamic properties of gaseous $\text{Mg}(\text{OH})_2$ have been presented in recent progress reports. Preliminary results on zirconia have been reported as has evidence for the existence of a gaseous uranium hydroxide.

Work This Period

Calculations have been completed on the vapor pressure of ZrO_2 . It is concluded that the vapor pressure of ZrO_2 is expressed by:

$$\log P_{\text{atm}} = -31,800/T + 6.98.$$

On the basis of this equation, the evaporation rate of zirconia would be 0.1 mil per year at 2000 K, and would be 0.01 mil per year at 1888 K (2939 F). Presence of hydrogen at 1 atm of pressure would increase the evaporation rate by a factor of approximately ten, but steam, per se, would not affect the vaporization rate.

Calculations concerning the enhancement in the volatility of uranium oxide by water vapor were completed. These calculations were based on the gaseous species being $\text{UO}_2(\text{OH})_2$. While no direct experimental proof can be given that this is the gaseous molecule, comparison with the tungsten- and molybdenum-oxygen-water systems would indicate that it is highly probable that this is a proper assumption. Further support for this model can be gained by the fact that the hydroxyl ion behaves quite similarly to the halide ions, its electronegativity being somewhere between fluorine and chlorine. (That stable gaseous uranyl halides form would indicate that it would be probable that a gaseous uranyl hydroxide would exist also.) The standard free energy of formation of gaseous $\text{UO}_2(\text{OH})_2$ can then be expressed as

$$\Delta G_f^\circ = -252,300 + 19.14 T \text{ kcal per mole}$$

from 1500 to 2000 K.

Attempts to determine experimentally the nature of a gaseous alumina species have not been successful since, if any such species is present, it is below the limit of detectability. It can be concluded that no aluminum-bearing species is present at a vapor pressure higher than 10^{-8} atm at 1800 K.

Plans for Future Work

Since zirconia has been found to be satisfactory in any atmosphere at temperatures to 3000 F (1922 K), major attention will be given to determining the effects of atmospheres on the stabilizers which are necessary if ZrO_2 is to be employed at elevated temperatures. Primary attention will be directed toward CaO and Y_2O_3 since these oxides are known to stabilize ZrO_2 and have relatively small cross sections for neutron absorption.

Thermal-Conductivity Studies

H. W. Deem and C. F. Lucks

Objective

To select a method and to design and assemble an apparatus for making thermal-conductivity (thermal-diffusivity) measurements on unirradiated and irradiated specimens containing coated-fuel particles.

Progress Approach

A survey of the literature indicated that a thermal-diffusivity method was the most practicable for the determination of the thermal conductivity of small irradiated specimens (1/4 in. in diameter by 0.080 in. thick) at elevated temperatures. In this method energy from a suitable light source, focused on the front surface of the specimen, is varied with time. The temperature response of the back surface of the specimen is measured, amplified, and displayed on an oscilloscope. Either the change in amplitude of the back-surface signal due to a change in frequency of the incident beam or the change in time response of the back surface due to a single incident pulse may be used to calculate the thermal diffusivity and thence the thermal conductivity of the specimen.

Progress Reported Previously

The basic apparatus has been assembled and tests run using the cyclic (variable frequency) method. This method proved unsatisfactory. Measurements made using the flash or single-pulse method with a laser beam as the light source proved satisfactory. Work has begun to modify the apparatus for the flash method, and to complete the apparatus for remote operation.

Work This Period

Forms for the 4-in.-thick lead shielding needed for experiments with irradiated specimens were completed. Lead was poured in the main form surrounding the vacuum chamber and this form is now complete and ready for installation. The cooling coils, vacuum port, and a mount for the exit mirror have been attached to the vacuum chamber, which is now ready for cleaning and assembly.

Plans for Future Work

The apparatus will be assembled and tests made to higher specimen temperatures. If these tests prove satisfactory, the apparatus will be moved to the hot cell and assembled for measurements on irradiated materials.

Evaluation of Commercial Products

H. S. Rosenberg, T. S. Elleman, and D. N. Sunderman

Objective

To provide information on the availability and quality of coated-fuel particles from commercial sources.

Program Approach

Interested manufacturers of coated particles who request information on the Coated-Particle Program are sent a standard form letter and summary of the evaluation procedures presently used for coated particles. Battelle is authorized to purchase 100-g lots of coated-fuel particles at a price of \$1 per g and to report the results of an evaluation of the material in this section of the report.

Progress Reported Previously

Requests for information on this program from 17 interested commercial firms have been received and answered. A 100-g lot of pyrolytic-carbon-coated natural UC_2 particles has been purchased from Minnesota Mining and Manufacturing Company. A 1-g sample of the as-received $3M$ particles has been neutron activated and submitted to postirradiation heating to measure the fission-gas release. The total xenon-133 release was 0.07 ppm after three successive 1-hr heat treatments at 815, 1090, and 1320 C.

Work This Period

No additional requests for information on this program were received during June.

Plans for Future Work

No future work is contemplated until other offers of commercial products are received.

Fission-Fragment-Induced Strain Studies

R. H. Barnes, T. S. Elleman, and D. N. Sunderman

Objective

To provide preliminary information for evaluating the extent to which mechanical strains are produced in ceramic-coating materials by recoiling fission fragments.

Program Approach

Flat thin sections of the material of interest with thicknesses of several fission-fragment ranges are irradiated in-pile with one face exposed to a source of recoiling fission fragments. Stresses generated in the side exposed to the fission recoils are expected to induce a permanent curvature in the thin section. Postirradiation measurements of the curvature of a thin section will then be employed to estimate the amount of strain produced by the fission recoils.

Progress Reported Previously

Six 0.5-in.-diameter alpha-alumina disks with thicknesses ranging from 5.5 to 7.7 mils were irradiated with one side of each disk facing a natural-uranium foil. The irradiation involved an exposure of 2 days in a thermal-neutron flux of about 1×10^{13} nv.

Work This Period

Surface-profile measurements were completed on both sides of each of the irradiated specimens. The profile measurements on the sides of the specimens not exposed to the fission recoils show profile changes which are thought to indicate that the fission-fragment irradiated sides of the specimens have expanded or swelled. The magnitudes of these changes are listed in Table L-7. The quantity listed is the change in specimen curvature which is caused by the irradiation where curvature is represented as the length of the line which can be drawn from the specimen center to a plane which passes through the edges of the specimen. In all measurements, the side opposite the one receiving the fission fragments became more concave following irradiation, supporting the view that the irradiated side expanded. These results show that the amount of change decreases with specimen thickness. The measurements of the changes are believed to have a standard deviation of about $\pm 8 \mu$. Profile measurements on the sides of the specimens exposed to recoils did indicate curvature changes; however, the changes were rather erratic and did not follow the logical pattern exhibited by the other side. This was probably caused by the variation of optical properties of the exposed surface and its influence on the profile measurements. Density determinations and surface studies on the specimens are now in progress.

TABLE L-7. MAXIMUM CHANGE OF SURFACE CURVATURE OF UNEXPOSED SURFACES OF ALUMINA DISKS EXPOSED TO FISSION RECOILS

Specimen Thickness, mils	Maximum Surface- Curvature Change ^(a) , μ
5.5	39
5.6	45
5.7	26
6.0	30
7.6	2
7.7	7

(a) Curvature is represented as the length of the line from the specimen center to the plane which intersects the specimen edges.

L-24

Plans for Future Work

Tentative plans have been made to perform an in-pile experiment where the fission-recoil-induced strains in alumina are measured continuously during irradiation.

CORROSION STUDIES OF THE FLUORIDE-VOLATILITY PROCESS (ORNL)

P. D. Miller, E. F. Stephan, and C. L. Peterson

Objective

To study the corrosion resistance of materials of construction for various components of the Fluoride-Volatility nuclear fuel-recovery process.

Program Approach

Candidate metals and alloys are being subjected to hydrofluorination and fluorination conditions in bench-scale equipment. These investigations are in support of nuclear fuel-reprocessing research being conducted by the Chemical Technology Division of ORNL.

Progress Reported Previously

Corrosion evaluations have shown HyMu-80 alloy to be a promising material of construction for fluorinators and hydrofluorinators. A previous progress report, BMI-1574, presented preliminary results for an alloy-development program aimed at even more resistant alloys. A continuing program involving alloys in the nickel-iron-molybdenum system was described last month in BMI-1581.

Work This Period

The alloys described in BMI-1581 have been rolled, cut, and heat treated. Equipment is being assembled to evaluate the corrosion resistance of the alloys.

Plans for Future Work

The specimens will be evaluated for resistance to hydrofluorinator conditions in the laboratory in a NaF-LiF-BeF₂ salt sparged with HF at 650 C.

PROGRESS ON USAEC/AECL COOPERATIVE PROGRAM (SRO)

S. W. Porembka

Work coordinated by SRO relating to the USAEC/AECL Cooperative Program is discussed in this section. Investigations concerning methods for end capping SAP-alloy fuel sheaths and cold joining of Zircaloy-2 and Type 410 stainless by roll forming and explosive welding have been concluded, and a topical report is being drafted summarizing progress in these areas.

Long-Term Creep-Rupture Program on SAP Alloys

J. A. VanECHO

Objective

To determine the effect of long-term exposure (to 20,000 hr) on the creep-rupture strength of three sintered aluminum-powder alloys (SAP-895, M-257, and a third yet to be determined) at 700, 800, and 900 F.

Program Approach

Test specimens are placed in standard Battelle creep frames and furnaces to determine their stress-rupture and creep strengths. Test temperatures are maintained to an accuracy of ± 2 F. Rupture times are recorded to the tenth of an hour by interruption of a timer when failure occurs. Creep measurements are taken daily or more often by means of an optical system which has a sensitivity of 0.00005 in. All requirements for creep testing specified by ASTM Standard E-139-58T are met or exceeded.

Stresses are chosen to cause failure in approximately 1, 10, 100, and 1000 hr on each material at each temperature. These data are then used to estimate stresses to cause failure in 10,000 and 100,000 hr. Actual tests will be made to verify the 10,000-hr stresses but the 100,000-hr tests will be discontinued at 20,000 hr. Creep data will be obtained from all tests.

Progress Reported Previously

All short-time creep-rupture tests had been completed on the SAP-895 alloy and two long-time (10,000 to 20,000 hr) tests were started at each test temperature (700, 800, and 900 F). Two tests were continued on the M-583 alloy at 700 and 800 F even though this alloy was removed from the program because of low strength and erratic behavior. Most of the preliminary tests (10 to 1000 hr) were completed on the M-257 alloy at 700 and 800 F, but none was started at 900 F. One long-time (10,000 hr) test was also in progress on this alloy at 800 F. Earlier in the program, extreme notch sensitivity of these alloys necessitated a redesign of the specimen and some changes in the test procedure. Results obtained to that time were considered invalid because of the notch failures and the program was essentially restarted. Additional material was needed to continue the program.

Work This Period

All of the creep-rupture tests reported in progress last month (BMI-1581) were continued through this period. These include six tests on the SAP-895 alloy, two on the M-583 alloy, and one on the M-257 alloy. All of these tests are expected to be 10,000 hr or longer in duration. Creep rates on these tests are relatively low and the increase in total deformation is generally very slight from one month to the next. Test results as of the end of this period are given in Tables Q-1, Q-2, and Q-3.

TABLE Q-1. CREEP-RUPTURE DATA ON SAP-895 SINTERED ALUMINUM-POWDER ALLOY AT 700, 800, AND 900 F

Specimen	Stress, psi	Rupture Time, hr	Elongation, per cent	Reduction of Area, per cent	Minimum Creep Rate, per cent per hr
<u>700 F</u>					
5-2	13,000	4.0	2.4	5.9	0.14
6-13	12,000	39.1	1.9	4.6	0.006
6-15	11,700	47.5	1.9	3.0	0.009
1-12	11,400	111.5	0.9	3.6	0.002
6-16	11,100	3148.9	1.8	6.1	0.000009
5-6	10,900	2038(a)	0.598	--	0.000015
5-8	10,700	813(a)	0.600	--	0.000080
<u>800 F</u>					
6-10	10,500	14.9	2.8	8.6	0.03
5-1	10,200	309.1	2.3	6.2	0.00025
6-8	10,000	1077.6	0.9	3.5	0.00005
5-7	9,750	2015(a)	0.620	--	0.000009
5-4	9,500	3309(a)	0.625	--	0.000007
<u>900 F</u>					
5-3	9,500	0.4	3.3	12.2	1.1
6-11	8,700	25.7	2.3	5.4	0.008
6-12	8,500	25.0	2.3	3.2	0.0035
6-9	8,000	259.4	2.8	7.0	0.00017
6-14	7,700	4365(a)	0.647	--	0.000002
5-5	7,500	2206(a)	0.540	--	0.000015

(a) Test in progress.

TABLE Q-2. CREEP-RUPTURE DATA ON M-583 SINTERED ALUMINUM-POWDER ALLOY AT 700, 800, AND 900 F

Specimen	Stress, psi	Rupture Time, hr	Elongation, per cent	Reduction of Area, per cent	Minimum Creep Rate, per cent per hr
<u>700 F</u>					
44-3	9,000	1.8	3.3	6.8	0.35
38-4	8,300	8.5	1.9	6.0	0.06
66-4	7,900	97.6	2.3	9.3	0.0023
15-1	7,700	119.9	2.4	4.1	0.0012
16-1	7,400	4005 ^(a)	0.904	--	0.00003
<u>800 F</u>					
246-4	7,000	19.2	2.3	10.7	0.03
44-4	6,500	19.2	2.8	6.2	0.023
38-3	6,300	224.1	2.3	8.5	0.0009
15-3	6,100	19.5	2.3	2.1	0.017
197-1	6,100	566.2	0.9	0	0.0004
197-2	5,600	3310 ^(a)	0.525	--	0.000007
<u>900 F</u>					
38-2	6,000	0.3	2.3	7.9	6.0
66-3	5,700	1.9	4.7	3.4	0.5
15-2	5,550	0.5	3.3	5.6	2.3
60-3	5,400	549.7	3.3	9.2	0.00001
15-4	5,250	1.5	2.8	4.5	0.75

(a) Test in progress.

TABLE Q-3. CREEP-RUPTURE DATA ON M-257 SINTERED ALUMINUM-POWDER ALLOY AT 700, 800, AND 900 F

Specimen	Stress, psi	Rupture Time, hr	Elongation, per cent	Reduction of Area, per cent	Minimum Creep Rate, per cent per hr
<u>700 F</u>					
1-4	11,600	22.4	1.8	3.6	0.03
1-3	11,000	43.5	3.3	14.9	0.013
75-1	11,000	54.7	3.7	8.8	0.013
1-5	10,500	139.6	2.8	5.2	0.0018
75-2	10,500	156.3	2.3	8.8	0.0015
1-6	10,000	2503.0	4.2	13.0	0.000020
2-6	9,750	335.3	3.3	--	0.0006
75-3	9,850	Scheduled	--	--	--
75-4	9,650	Scheduled	--	--	--
<u>800 F</u>					
2-1	10,500	0.5	3.3	10.0	1.3
2-2	9,200	5.4	2.8	6.1	0.11
2-3	8,700	30.4	2.3	6.1	0.010
714-1	8,500	21.0	4.2	15.5	0.027
76-1	8,500	14.5	4.2	9.9	--
2-4	8,400	79.2	3.3	9.0	0.0018
76-2	8,200	223.9	2.8	6.1	0.0008
2-5	7,900	3621(a)	0.898	--	0.000018
76-3	7,700	335(a)	0.618	--	0.00025
<u>900 F</u>					
78-2	10,000	0.01	7.0	32.9	--
78-2	7,000	2.0	3.8	8.1	0.60
714-2	6,300	195(a)	0.775	--	0.0007
78-3	6,000	432(a)	0.665	--	0.00012

(a) Test in progress.

Additional M-257 material was received and machined into specimens. A number of short-time tests were made on this alloy at all three test temperatures. Check tests showed this particular batch of material to have essentially the same creep-rupture strength as the material already in test. In addition, two long-time tests are scheduled to start at 700 F and one more at 800 F. Since no tests had yet been made at 900 F, the short-time tests are still in progress at this temperature.

Plans for Future Work

The tests now in progress will be continued until failure or until they have reached their scheduled termination. All tests on the SAP-895 and the M-583 alloys are scheduled to run 10,000 or 20,000 hr. This is also true for the tests in progress and scheduled on the M-257 alloy at 700 and 800 F. The only short-time tests remaining are on the M-257 alloy at 900 F. As soon as these are completed, two long-time tests will be started at this temperature.

The program, in general, remains unchanged. It consists, essentially, of making short-time creep-rupture tests to establish the effect of exposure to times up to about 1000 hr. Once this is completed, the results will be used to determine stresses for rupture in 10,000 and 100,000 hr for each alloy at each test temperature.

One more alloy remains to be chosen for this program. A similar series of tests will be initiated as soon as it is received.

FISSION-PRODUCT DEPOSITION STUDIES (ORNL)

W. G. Rieder, R. A. Ewing, M. S. Edwards, and G. E. Raines

Objective

To study and correlate the deposition behavior of various fission products in an out-of-pile loop system.

Program Approach

Deposition behavior is to be studied by a series of out-of-pile experiments which will consist of installing a mildly irradiated fuel specimen in a recirculating gas loop; circulating the gas at prescribed flow and temperature conditions past the specimen continuously for 2 to 4 days; monitoring gross activity buildup at different locations as a function of time; and, at the conclusion of the run, dismantling the loop and radiochemically analyzing for various deposits.

The experimentally determined deposition patterns will be characterized by both empirical, and insofar as possible, theoretical correlations. These correlations should consider Reynolds number, velocity, surface condition, material of construction, geometry (including orifices, etc.), particulate matter in the gas stream, fission-product species (and form of the fission product - elemental, carbide, oxide, etc.), and its diffusion coefficient.

Uranium dicarbide will be employed as the fuel material. Initial experiments will be conducted with helium, although it is anticipated that some later experiments may be conducted in the same facility utilizing argon so that higher Reynolds-number ranges may be explored.

Progress Reported Previously

This is the initial report on the program.

Work This Period

Design of the experimental facility and fabrication of component parts were completed. The main flow passage of the loop consists of 1/2-in. -OD 16-gage Type 304 stainless steel tubing. All instruments and vendor-furnished equipment were obtained except the major part of the flow-control system. Assembly of the apparatus and instrumentation console is well under way. The system is designed for the following minimum performance capability:

Reynolds number	10,000
Maximum gas temperature	1200 F
Fuel-specimen temperature	1800 F
Maximum gas temperature in blower	500 F

These conditions are to be achieved simultaneously so that exchange of one condition for another will be possible; e.g., higher Reynolds numbers will be investigated by increasing the velocity, but the temperature drop and maximum gas temperature will be reduced during these runs.

A number of fuel specimens were prepared; no evaluation of these is yet available. The configuration chosen is a hollow graphite cylinder (about 0.75 in. in OD by 0.37 in. in ID by 0.5 in. long) which contains fully enriched UC_2 (about 100 mg) particles about 100 μ in diameter with a 4- μ dense carbon coating on each individual particle. The ID of the cylinder corresponds to the flow passage in the loop.

A theoretical parameter-variation study was conducted utilizing the Bendix G-20 digital computer. A finite difference approximation to the diffusion equation allowing for flow and utilizing cylindrical coordinates was employed. The results have not been appraised. One problem was also solved utilizing resistance-network analog techniques for comparison.

R-2

Plans for Future Work

The assembly of the apparatus will be completed, the results of the parameter study will be appraised, and a series of experiments will be conducted with primary consideration to the establishment of Reynolds-number, velocity, and geometry effects.

RADIATION-EFFECTS STUDY OF CANDIDATE FUEL MATERIALS FOR THE MGCR (GA)

P. Steinback, J. E. Gates, and W. H. Goldthwaite

Objective

To determine the irradiation stability of selected ceramic-matrix fuels under conditions of temperature and burnup similar to those anticipated in the MGCR.

Program Approach

Battelle has assisted General Atomic in the development and evaluation of core materials for the Maritime Gas-Cooled Reactor and the High-Temperature Gas Reactor, and is now participating in studies of the irradiation damage of BeO-matrix fuel up to relatively high burnups. Currently, Hastelloy X-canned BeO-UO₂ fuel pellets are being irradiated in a capsule which is equipped with thermocouples and auxiliary electric heaters and is capable of maintaining fuel pellet-surface temperatures in the 1500 to 1800 F range, primarily by fission heating, for an irradiation as long as 18 calendar-months. At the present time, this capsule (BMI-36-1) is scheduled to be irradiated for about 1 year. Target burnup of the uranium-235 is 25 to 35 per cent. Data concerning specimen burnup, swelling, fission-gas release, and microstructural changes are to be obtained in postirradiation hot-cell examination.

Progress Reported Previously

Capsule BMI-36-1, containing 12 fuel pellets of 30 volume per cent UO₂ (12 per cent enriched) in BeO placed in six Hastelloy X can compartments, was prepared for irradiation in the MTR. The irradiation began with MTR Cycle 165 on November 10, 1961, in a specimen-effective peak flux of 2×10^{13} nv. Vertical relocation of the capsule within its facility has been employed to place the canned fuel pellets more nearly across the peak of the thermal-neutron flux profile. Fuel pellet-surface temperatures during this irradiation have ranged from approximately 1410 to 1750 F, as indicated by six nearby reference thermocouples.

On May 3, a previously satisfactory thermocouple, that in the topmost reference position, began to exhibit occasional periods of open circuit. On May 28 the five other thermocouples were still operating satisfactorily. The two auxiliary electric heaters remaining in use were still operating satisfactorily.

Work This Period

The irradiation of Capsule BMI-36-1 was continued. During the period May 31 through June 25 (encompassing the last part of Cycle 174 and all of Cycle 175), fuel pellet-surface temperatures ranged from 1510 to 1700 F. Since June 6, both the topmost and bottommost reference thermocouples have exhibited recurrent malfunction. The thermocouple immediately above the lowest showed a 100 F drop in its indicated temperature on June 20 and continues to indicate that lower temperature. The three other thermocouples and the two auxiliary electric heaters being employed all remain satisfactory. A third heater, which shows excessive leakage current, continues to be withheld from service.

The estimated accumulated burnup of uranium-235 in the fuel pellets for the first ten irradiation cycles (Cycle 165 through 174, November 10, 1961, through June 4, 1962) is 18.0 per cent. The burnup through June 25 (through Cycle 175) is about 20 per cent.

Plans for Future Work

During the reactor shutdown before Cycle 177 (about July 16), Capsule BMI-36-1 will be lowered 2 in. within its facility to improve the neutron irradiation and temperature profiles. During the shutdown before Cycle 178 or a shutdown thereafter, the capsule will be moved to another reactor position having a slightly higher thermal-neutron flux. This move will compensate for capsule fuel burnup and decrease the demand on the auxiliary electric heaters. The auxiliary electric heater which was taken out of service will be turned on periodically, and the leakage currents will be recorded. The irradiation will be continued.

GAS-COOLED REACTOR PROGRAM (AGN)

D. L. Keller

The Army Reactor Branch (ARB) is developing small portable reactor systems for military applications. Aerojet-General Nucleonics (AGN) is the prime contractor under the Army Gas-Cooled Reactor Systems Program (AGCRSP) and is charged with the design, construction, and operation of the GCRE and ML-1. The studies below are in support of the ML-1 program.

ZrO₂-UO₂ Fabrication Studies

S. J. Paprocki, D. L. Keller, J. M. Fackelmann, and T. R. Wright

Objective

To study fabrication parameters and phase relationships in the UO₂-ZrO₂ system, and the radiation stability of such bodies.

Program Approach

Bodies of UO₂-ZrO₂ will be fabricated by mixing the individual powders and sintering. The effect of such variables as powder particle size, time, temperature, and atmosphere on the resultant density and constitution on such bodies will then be studied. Metallographic and X-ray diffraction techniques will be used to study these variables, with primary emphasis being on compositions in the vicinity of UO₂-50 mole per cent ZrO₂.

Progress Reported Previously

Vacuum sintering mixtures of UO₂ with 11, 59, and 87 mole per cent ZrO₂ at 1700 C (2 hr) and 1800 C (3 hr) failed to densify the compacts. The structures of the 11 and 59 mole per cent ZrO₂ specimens appeared homogeneous metallographically; however, very fine ZrO₂-rich particles may have been present. The 87 mole per cent ZrO₂ showed some original UO₂ particles to be quite low in zirconium.

Work This Period

Hydrostatically pressed pellets of UO₂ with 0, 3, 15, 50, and 100 mole per cent ZrO₂ were sintered in hydrogen for 20 hr at 1700 C. Densities of the compacts before and after sintering are given in Table U-1. As may be seen, sintered densities varied from 84 to 94 per cent of theoretical, as determined from measuring and weighing the compacts.

X-ray diffraction analysis of the 3, 15, and 50 mole per cent mixtures showed line broadening in the 50 mole per cent mixture and clear, narrow lines in the other two mixes corresponding to fcc-phase lattice constants of 5.4601 ± 0.0001 A and 5.4232 ± 0.0003 A, respectively. The line broadening in the 50 mole per cent alloy was probably caused by slow cooling through the high-temperature two-phase region.

The powders used in these experiments were Mallinckrodt ceramic-grade UO₂ having a nominal average particle size of 9 μ and Baker Chemical Company ZrO₂. Mixing was accomplished by ball-milling the two powders 24 hr in water-filled steel ball mills. Spectrographic analyses showed that 0.1 to 1 w/o iron was present after milling compared with 40 ppm in the starting powder.

TABLE U-1. DENSITY OF COMPACTS OF $\text{UO}_2\text{-ZrO}_2$ BALL-MILLED MIXTURES
AFTER SINTERING 20 HR AT 1700 C IN HYDROGEN

ZrO ₂ , mole per cent	Density, g per cm ³		Sintered Density, per cent of theoretical
	Green	Sintered	
0(a)	7.3	9.2	84
0	7.1	10.2	93
3	6.8	10.1	94
15	7.0	9.3	91
50	5.3	7.6	91
100	3.5	--	--

(a) As-received 9- μ powder used without ball milling.

Coulter counter measurements of powder particle sizes before and after ball milling showed mean particle sizes of 2.5, 4.5, 8 μ for ball-milled UO_2 , as-received ZrO_2 , and as-received UO_2 , respectively.

Plans for Future Work

Spectrographic analyses will be conducted on the mixtures from the stainless steel millings to determine the amount of iron pickup. Millings are planned using a rubber-lined mill and uranium balls to determine the most effective grain-size reduction procedure with the least impurity pickup. Compaction studies will be conducted to determine optimum forming pressures. Sintering studies will be conducted in hydrogen and steam atmospheres in an attempt to densify the compacts. Dense homogeneous bodies will be irradiated under ML-1 conditions.

Once successful fabrication of the compacts has been accomplished, a series of compacts, prepared in the optimum manner, will be subjected to X-ray and metallographic analyses to determine the phase relationships in the $\text{UO}_2\text{-ZrO}_2$ system.

ZrO₂-UO₂ Design Studies

F. Young, W. S. Hogan, and D. Dingee

Objective

To investigate possible effects, relative to the reactor physics of the ML-1 reactor, which might result from replacing the BeO-UO_2 portion of the fuel with $\text{ZrO}_2\text{-UO}_2$.

Program Approach

The $\text{ZrO}_2\text{-UO}_2$ offers a possible increase in core-life because of a higher burnup potential. The objective was to investigate such a fuel diluent replacement with respect to the design and safety of the ML-1 reactor. The program was divided into the following phases:

- (1) A review of ML-1 design and safety, through quarterly progress reports, to help determine in what areas the fuel materials properties may significantly affect the reactor properties of interest
- (2) A discussion with appropriate personnel from AGN to obtain information and clarification relative to this problem
- (3) Performance of such studies as were indicated from Phases (1) and (2).

Progress Reported Previously

This is the only monthly progress report on this work.

Work This Period

Because the ML-1 reactor core is quite heterogeneous, difficulties were encountered in applying calculational methods directly to this assembly. For this reason, the core was idealized to a large degree, and one-group diffusion theory was utilized for the calculation of lattice parameters and temperature distributions. With these approximations in mind, the calculations were done on a relative basis since it was impractical to rely on the accuracy obtained in an absolute sense; thus, for the determination of each parameter with ZrO_2 as a diluent, the same determination was made with the reference BeO diluent, and the fractional deviation of each parameter was used as a basis for judgement. The following assumptions were made as a basis of calculation:

- (1) Reactor thermal power remains unchanged
 - (2) Fuel-to-diluent volumetric ratio remains unchanged
 - (3) Fuel-element design remains unchanged except for substitution of ZrO_2 for BeO .
- Calculations yield the following:

- (1) The infinite multiplication factor changed less than 1 per cent
- (2) The maximum cladding temperature remained the same.
- (3) The maximum fuel center-line temperature increased by approximately 900 F for UO_2 - ZrO_2 pins; it remained the same for UO_2 pins.

The fact that the infinite multiplication factor remains essentially constant reflects the fact that the volume-weighted macroscopic cross section of the diluent is negligible when compared to that of the UO_2 . There will be a slight decrease in the finite multiplication factor because of the smaller slowing down power of ZrO_2 as compared with BeO , which should increase fast leakage. This change should be extremely small since the volume ratio of BeO to moderator is only 0.056.

The increase in center-line temperature results from the lower thermal conductivity of ZrO_2 as compared with BeO . This parameter was evaluated for the composite pellets by utilizing Maxwell's formula for two-phase mixtures.

Because of increased core life, a control problem was expected. That is, the same process of shim and poison management was not expected to be optimum for both short and extended core-life expectancies. However, because of the small change in the multiplication factor, this problem is considered to be the same as that which would result in the BeO - UO_2 -fueled core if greater burnups were feasible. This has been considered in IDO-28581. Experiments reported there indicate that shims should be removed after about 4000 hr of full-power operation to continue the operation. This same manipulation should suffice to continue operation with the ZrO_2 - UO_2 -fueled core after a comparable time of operation.

Based upon the brief investigations in the areas indicated no major reactor safety or design problems are foreseen in substituting ZrO_2 for BeO as a fuel diluent in the ML-1 reactor.

Plans for Future Work

No future studies are planned on this program.

Postirradiation Examination of Hastelloy X Cladding From AGN 1B-8T-1 Subassembly

J. B. Melehan, D. A. Vaughan, and J. H. Saling

Objective

To examine and evaluate Hastelloy X tubing which served as cladding for UO_2 and UO_2 - BeO fuels during irradiation.

Program Approach

The AGN 1B-8T-1 subassembly consisted of UO_2 and 70 w/o UO_2 -BeO fuels enclosed in Hastelloy X cladding tubes. The subassembly was irradiated at the MTR in a nitrogen-0.5 volume per cent oxygen atmosphere at a maximum cladding-surface temperature of 1750 F. Preliminary postirradiation examination of the fuel pins indicated embrittlement of the cladding. Metallographic examination at the MTR laboratories revealed microstructural modification of the cladding. The typical preirradiation microstructure of Hastelloy X is characterized by finely divided precipitate particles in a single-phase matrix. In the postirradiation microstructure an extensive second phase was observed, located principally in the grain boundaries.

The present program will include the following study of the irradiated cladding material:

- (1) Determination of tensile properties (yield strength and elongation)
- (2) Measurement of microhardness
- (3) Metallographic examination and phase identification
- (4) Determination of chromium composition gradient
- (5) Determination of oxygen, carbon, and nitrogen
- (6) X-ray diffraction and electron probe microanalysis.

Additional investigations concerned with unirradiated control materials will include:

- (1) Determination of tensile properties of Hastelloy X in the as-received condition and following a 5000-hr exposure at 1750 F in the nitrogen-0.5 volume per cent oxygen atmosphere.
- (2) Development of techniques for detecting chromium depletion.
- (3) Metallographic phase identification on unirradiated Hastelloy X.

Progress Reported Previously

Work on the program has just been initiated.

Work This Period

Irradiated fuel pins, both intact and already sectioned, were received at the BMI Hot-Cell Facility. In all cases the pins were bent; this will result in eccentric loading during tensile testing. Each full-length pin was cut into four approximately equal pieces about 6 in. long. These sections were immersed in a boiling solution of the following composition for removal of the UO_2 and UO_2 -BeO fuel:

HNO_3	87 cm ³
NaF	0.168 g
$\text{Al}(\text{NO}_3)_3$	1.044 g
H_2O	13 cm ³

There was no obvious indication of attack on the Hastelloy X.

Additional samples of as-received Hastelloy X tube were received from AGN. Several samples of Hastelloy X tubing following heat treatment in nitrogen-0.5 volume per cent oxygen at 1750 F for 5000 hr were available at the BMI fatigue-test laboratory.

The several specimens described in Table U-2 were available in the unirradiated condition for the development of analytical techniques in metallography, including microchemical analysis, X-ray diffraction, and electron-probe microanalysis. The results available to date are shown in Table U-2.

TABLE U-2. DESCRIPTION OF HASTELLOY X SPECIMENS EMPLOYED IN TECHNIQUE DEVELOPMENT

Specimen	Heat	Heat Treatment	Observations	
			Metallography	X-Ray
6A	4762	2500 hr at 1750 F in nitrogen	Continuous grain-boundary phase; large intragranular precipitate; staining shows high chromium content in grain-boundary phase	M_6C , $a_0 \approx 10.82 \text{ \AA}$, Phase X
7A		2500 hr at 1750 F in argon	Discontinuous grain-boundary phase plus an intragranular precipitate	M_6C , $a_0 \approx 10.95 \text{ \AA}$
8A	4762	500 hr at 1750 F in argon	No work to date	No work to date
9A		2500 hr at 1750 F in argon	No work to date	No work to date
10A	E9517	5000 hr at 1750 F in $N_2 + 0.5\% O_2$ followed by fatigue test for 45 hr at 1400 F in air; specimen from section in clamp	Discontinuous grain-boundary phase plus an intragranular precipitate	M_6C , $a_0 \approx 10.95 \text{ \AA}$
11A		5000 hr at 1750 F in $N_2 + 0.5\% O_2$ (Center of Pin 13)	Very narrow discontinuous grain-boundary phase plus a small amount of intragranular precipitate	M_6C , $a_0 \approx 10.95 \text{ \AA}$

The microstructure of Specimen 6A is extremely interesting in that it is very similar to that seen in the brittle portion of Pin 12 examined at Phillips Petroleum in which a continuous grain-boundary phase was observed. In Specimen 6A a new phase (X) was detected by X-ray diffraction and identified as the grain-boundary phase. Chemical staining revealed this phase to be very high in chromium. Although its pattern is similar to sigma, a positive identification has not been made. The intragranular precipitate has been identified as an M_6C type, but its metal composition has not been determined. The low lattice parameter in comparison with the other specimens suggests some variations. It is believed that the nitrogen gas has modified the diffusion behavior in Hastelloy X even though some oxidation occurred as evidenced by the presence of Cr_2O_3 and $MnCr_2O_4$ on the surface of Specimen 6A. It is interesting to note that heat treatment in an inert gas (argon) did not result in a heavy growth of the grain-boundary phase, suggesting the driving force for diffusion was the presence of the nitrogen and/or oxygen.

The marked difference between Specimens 10A and 11A is surprising in that a heat treatment for 45 hr at 1400 C modified the distribution of the precipitate significantly. This may be due to the change in atmosphere during the fatigue test.

Plans for Future Work

The postirradiation studies will continue with tensile testing of irradiated cladding. Tensile tests will also be performed on unirradiated control materials. The efforts will continue at devising techniques for postirradiation X-ray diffraction analysis, microprobe analysis, and chromium-depletion detection.

PROCESS DEVELOPMENT AND EVALUATION OF PROPERTIES OF F-48 NIOBIUM ALLOY (LRL)

K. R. Grube and E. S. Bartlett

Objective

To obtain data on the strength, thermal expansion, and thermal conductivity of F-48 alloy, and to determine variations in elevated-temperature strength from heat to heat and as a result of fabrication variables.

Program Approach

Sections of F-48 alloy plates produced by a commercial supplier are submitted to Battelle for evaluation. Tensile, creep, impact, thermal-expansion, and thermal-conductivity measurements, and chemical analyses constitute the evaluation program.

A special study to evaluate the effects of ingot size will examine only mechanical properties, microstructure, and chemistry.

Progress Reported Previously

Three plates from one heat of F-48 alloy were received in the stress-relieved condition and evaluated for tensile, creep, and impact properties. Microexamination indicated the plates to be partially recrystallized.

Low ductility (<10 per cent elongation) was observed at room temperature. Tensile and yield strength decreased, and ductility increased with temperature. Strain-aging occurred between 700 and 1600 F. Direction of specimen, relative to fabrication direction, had very little effect on tensile properties. Impact transition temperature (temperature for one-half of total energy decrease) for F-48 alloy ranged from 515 to 565 F, depending upon direction relative to prior fabrication.

Work This Period

Tensile and impact transition temperatures were determined for two plates of F-48 alloy fabricated from 6- and 8-in. -diameter ingots. Material from the 8-in. ingot exhibited slightly better over-all ductile-to-brittle tensile transition behavior, although material from the 6-in. ingot retained slightly superior ductility in the longitudinal direction at intermediate temperatures (125 to 300 F).

Similarly, impact transition behavior was modestly superior in material fabricated from the 8-in. ingots.

Plans for Future Work

Thermal-conductivity measurements, creep-property determination, and the statistical analyses will be completed during the next work period.

GAS-PRESSURE BONDING OF BERYLLIUM-CLAD ELEMENTS (ORNL)

S. J. Paprocki, E. S. Hodge, H. D. Hanes, and G. R. Goetsch

Objective

To determine the conditions necessary for the self-bonding of beryllium and the fabrication techniques for cladding UO_2 fuel-core platelets with beryllium.

Program Approach

This work is in support of ORNL's studies on potential materials for gas-cooled reactors. The gas-pressure-bonding technique is being used to fabricate flat-plate specimens containing four UO_2 cores measuring 4 by 1 by 1/4 in. Evaluation is based upon bond and cladding properties after bonding as determined by metallographic examination and mechanical testing.

Progress Previously Reported

The self-bonding studies conducted with beryllium showed that the best bond was achieved by grit blasting the mating surfaces prior to bonding. Grain growth across these interfaces was in excess of 50 per cent. Other methods of preparing the bonding surfaces such as abrading, shapering, and the use of powder as a surface activator were not as successful.

The UO_2 cores coated with vapor-deposited chromium showed no reaction between core and beryllium cladding.

Beryllia (BeO) was shown to be a satisfactory barrier material to prevent reaction between the cladding and the spacer materials used during bonding operation if the coating is of sufficient thickness and is uniform. However, it does not provide a satisfactory surface on the beryllium after removal of the spacer.

Bonds achieved by bonding at 1550 F were just as good as those made at 1650 F.

The cracks in the cladding of the beryllium fuel elements are not due entirely to the decanning process nor to the isostatic properties of the beryllium sheet used to fabricate the fuel-element components.

Work This Period

Three tensile specimens were made with a bond interface at the midpoint of the length of the specimen. This was done by assembling, end-to-end, two 1-in. -diameter rods 2 in. long in a mild-steel tubular can. They were then gas-pressure bonded at 1650 F for 4 hr at 10,000 psi.

The results of the tensile test were favorable in that all three required a stress over 35,000 psi to cause failure, and two of the specimens fractured in the parent material rather than at the bond interface.

Plans for Future Work

The final report is in the process of being written.

RWD/RFD:all